Spin ladders and quantum simulators for Tomonaga–Luttinger liquids

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Abstract

Magnetic insulators have proven to be usable as quantum simulators for itinerant interacting quantum systems. In particular the compound (C\textsubscript{5}H\textsubscript{12}N\textsubscript{2})\textsubscript{2}CuBr\textsubscript{4} (for short: (Hpip\textsubscript{2})\textsubscript{2}CuBr\textsubscript{4}) was shown to be a remarkable realization of a Tomonaga–Luttinger liquid (TLL) and allowed us to quantitatively test the TLL theory. Substitution weakly disorders this class of compounds and thus allows us to use them to tackle questions pertaining to the effect of disorder in TLL as well, such as that of the formation of the Bose glass. In this paper we present, as a first step in this direction, a study of the properties of the related (Hpip\textsubscript{2})\textsubscript{2}CuCl\textsubscript{4} compound. We determine the exchange couplings and compute the temperature and magnetic field dependence of the specific heat, using a finite temperature density matrix renormalization group procedure. Comparison with the measured specific heat at zero magnetic field confirms the exchange parameters and Hamiltonian for the (Hpip\textsubscript{2})\textsubscript{2}CuCl\textsubscript{4} compound, giving the basis needed to begin studying the disorder effects.

(Some figures may appear in colour only in the online journal)

1. Introduction

Understanding the physics of quantum interacting systems is one of the most challenging problems of condensed matter physics. This is especially true for low dimensions where the interaction effects are usually reinforced by the dimensional confinement and lead to novel physics. In particular, in one dimension, all the low energy excitations become collective ones and the usual Landau or Bogoliubov quasiparticles [1] do not exist. The resulting physics, which presents a unique set of universal properties, can be described by the Tomonaga–Luttinger liquid (TLL) theory, characterized by power law decay of the correlation functions and fractionalization of the excitations [2].

TLL physics manifests itself in many different experimental situations, which are the object of this special issue, and in particular the power law behavior of the correlations has been observed in several situations [3–6] (see also [2, 7] for additional references). However for many of the experimental systems, it is difficult to go beyond the actual observation of the power law behavior, and fully test the TLL theory. Indeed, in the condensed matter context, the interactions are usually poorly known (typically a screened Coulomb interaction), which makes an \textit{ab initio} calculation of the TLL exponent impossible. In addition it is usually difficult to test the predicted universality of the TLL behavior (namely that all the exponents are functionally related to a unique parameter $K$ [2]) since one often lacks a control parameter or
the possibility of easily accessing several correlation functions simultaneously.

As a result, it is highly desirable to dispose of ‘quantum simulators’ to realize TLLs, namely experimental systems that can be faithfully described by a simple model Hamiltonian and for which quantitative comparison with the experiments is possible. Prime candidates for such quantum simulators are provided by cold atomic systems [8]. Their remarkable versatility and degree of control of the dimensionality of the problem, of the kinetic energy and of the interactions has made them invaluable tools for tackling several properties of strongly correlated systems. However for TLL physics, and despite remarkable success in realizing and probing one-dimensional systems, they still suffer from limitations coming from either the confining potential, that corresponds to a space varying chemical potential and thus blurs the exponents, or limitations of interactions for the systems without the confining potential (see e.g. [9, 10]). Another class of quantum simulators that has proven very successful is that of the magnetic insulators [11]. Indeed such spin systems can be mapped onto interacting boson systems. The density of bosons can be controlled by a magnetic field and easily measured from the magnetization. In high dimensions, they have proven very successful for studying effects such as the Bose–Einstein condensation [12–14]. In one dimension these systems provide a remarkable realization of a TLL. Because the ‘interactions’ between the ‘bosons’ are now provided by the magnetic exchanges, they are very well known and short range, allowing a direct calculation of the TLL parameters without any fudging. Comparison between such calculations and experimental results on the metal–organic spin ladder [15] piperidinium copper bromide (C₃H₁₂N)₂CuBr₄, for short (Hpip)₂CuBr₄, have provided the first quantitative test of the TLL theory [16–19]. Since these first experiments, the successful comparison between theory and experiment in this class of compounds has been extended to more refined correlations such as the ones measured in electron spin resonance (ESR [20]), or neutron scattering experiments [19, 21]. Other classes of ladder compounds have also been investigated [22–24].

Among the interesting effects that could be studied with such a class of material are the effects of disorder on interacting quantum particles. Indeed, disordered bosons lead to interesting phases such as the Bose glass phase, predicted a long time ago [25, 26], whose experimental realization is still elusive. Magnetic insulators provide a very nice playground for such effects, since chemical substitution allows us to weakly affect the spin exchange and thus to introduce a weak disorder in the bosons. Several materials have been exploited to test for the presence of a Bose glass phase in various dimensions—ranging from three-dimensional to quasi-one-dimensional systems [11, 27–29]. Given the importance of the (Hpip)₂CuBr₄ compound and the excellent realization of one-dimensional TLL that it offers, it is particularly important to be able to control the disorder effects in this material.

We thus introduce in this paper the disordered version (Hpip)₂CuBr₄(1–x)Clₓ, in which the Br atoms have been replaced partially by Cl, and studies of the parent compounds (Hpip)₂CuBr₄ and (Hpip)₂CuCl₄. We investigate various quantities such as the specific heat and show how one can extract the various characteristic parameters of the system.

The plan of the paper is as follows. In section 2 we present the general properties of the compounds and their theoretical description. In section 3 we present the results for the specific heat measurements and the comparison with the density matrix renormalization group (DMRG) calculations, as well as a description of the methodology. In section 4 we discuss some of the consequences and perspectives for this class of compound.

### 2. Generalities on (Hpip)₂CuBr₄

#### 2.1. The compound

Recent successes in the synthesis and the growth of single crystals of new metal–organic compounds have opened up exciting new routes for experimental studies of model magnetic materials. This is due to the typical energy scale of the magnetic exchange interactions in such materials, which are on the order of meV, and exceptionally clean realizations of some low dimensional exchange geometries, e.g. one-dimensional spin ladders and two-dimensional square lattices.

In (Hpip)₂CuBr₄ the magnetic Cu²⁺ ions with quantum spin \( S = 1/2 \) form one-dimensional ladder-like structural units. Magnetic exchange interactions in these ladder arrays are via Cu–Br–Br–Cu super-exchange paths. As demonstrated in figure 1, two such paths contribute to the exchange on ladder rungs (\( J_\perp \)), whereas one, longer path constitutes the ladder leg (\( J_\parallel \)). The possible interladder exchange (\( J' \)) is very small due to the large organic (C₃H₁₂N)+ piperidinium ion effectively separating the ladder units.

While only experiments using neutron inelastic scattering are able to unambiguously determine the exchange Hamiltonian of such a spin system [21], additional measurements of bulk magnetic properties, such as the uniform magnetization with clear square-root field dependences near the critical magnetic fields, may indicate the excellent low dimensionality of a material [15, 16]. Once the ladder Hamiltonian is confirmed, the critical magnetic fields \( h_{c1} \) and \( h_{c2} \) are used to extract precise values for \( J_\| \) and \( J_\perp \) as will be described below.

The exchange parameters for (Hpip)₂CuBr₄, as summarized in table 1, place the material in the so-called strong-coupling limit of the quantum spin ladder. The corresponding, relatively low critical fields for the first time allowed experimental studies of all its TLL properties by a

| Compound            | \( J_\parallel \) (K) | \( J_\perp \) (K) | \( h_{c1} \) (T) | \( h_{c2} \) (T) |
|---------------------|----------------------|-------------------|-----------------|-----------------|
| (Hpip)₂CuBr₄        | 3.52                 | 1.13              | 1.8             | 4.9             |
| (Hpip)₂CuCl₄        | 5.10                 | 3.06              | 2.4             | 20              |
| (Hpip)₂CuBr₄Cl₂     | 12.6                 | 3.55              | 6.73            | 13.79           |
| (Hpip)₂CuCl₄Cl₂     | 3.52                 | 1.13              | 1.8             | 4.9             |

Table 1. Summary of ladder exchange parameters and critical fields from uniform magnetization data.
exchange paths mediated by Br⁻ sites. Since the Br/Cl site affects the super-exchange paths, one can thus expect a modification of the value of the exchange parameters. Such modification is indicated in figure 1. The parameters, extracted previously from magnetization data, for three compounds are summarized in table 1.

We will show in the following sections how one can characterize the compound (Hpip)₂CuCl₄, namely ascertain the structure of its Hamiltonian as well as determine its parameters.

2.2. The theoretical description

The piperidinium compounds are very well described by a spin ladder Heisenberg Hamiltonian

\[ H_\mu = J_\perp H_\perp + J_\parallel H_\parallel \]  

where

\[ H_\perp = \sum_{l=1}^{L} S_{l,1} \cdot S_{l,2} - \hbar^2 J_{\perp}^{-1} M^2 \]  

\[ H_\parallel = \sum_{k=1,2} \sum_{l=1}^{L-1} S_{l,k} \cdot S_{l+1,k} \]

The magnetic field, \( \hbar^2 \), is applied in the z direction, and \( M^2 \) is the \( z \)-component of the total spin operator \( \mathbf{M} = \sum_{l=1}^{L} (S_{l,1} + S_{l,2}) \). The operator \( S_{l,k} = (S_{l,k}^x, S_{l,k}^y, S_{l,k}^z) \) acts at the site \( l \) (\( i = 1, 2, \ldots, L \)) of the leg \( k \) (\( k = 1, 2 \)). The \( S_{l,k}^\alpha \) (\( \alpha = x, y, z \)) are conventional spin-1/2 operators, \( [S_{l,k}^x, S_{l,k}^y] = iS_{l,k}^z \), and \( S_{l,k}^{++} = S_{l,k}^+ \cdot S_{l,k}^- \). In addition, there is a weak interladder coupling that we will not consider here (for more details on its effects see [19]). The magnetic field in teslas is related to \( h^2 \) with \( \mu_B \) being the Bohr magneton and \( g \) being the Landé factor of the unpaired copper electron spins.

ESR measurements have confirmed that anisotropies are only of the order of a few per cent [31, 20].

In the absence of a magnetic field, the ground state of the system is a superposition of dimers, in a singlet state, separated by a gap of order \( J_\parallel \) from the triply degenerate excited triplet states. Application of the magnetic field reduces the energy of one of the triplets until it crosses the level of the singlet. Since the triplet can delocalize from rung to rung because of the exchange \( J_\parallel \), we have a band of triplets. We thus have two quantum phase transitions. At \( h_{c1} \) the first triplet (from the bottom of the band of triplets) enters the system, while at \( h_{c2} \) the triplet band is full. The triplets can be faithfully represented by hard core bosons, or in one dimension by spinless fermions using the Jordan–Wigner transformation. The magnetic field is thus acting as a ‘gate voltage’ for an electronic system controlling the chemical potential of the triplets. The density of triplets is directly measured by the magnetization of the system which will increase from zero to one per rung. The triplets are of course interacting due to the magnetic exchange. As indicated in figure 3, the system has three phases. For \( h < h_{c1} \) it is in a gapped state with essentially a ground state made of singlets. For \( h_{c1} < h < h_{c2} \) it contains interacting triplets. It is thus

number of high precision experimental techniques [15–18, 21].

In addition to the nearly optimal spin ladder properties of this compound, its chemical flexibility can be explored to realize the full potential of such metal–organic compounds as low dimensional model systems, from which further aspects of TLL can be studied in great detail. Full and partial substitution of the Br by Cl has been demonstrated; see figure 2. The chloride (Hpip)₂CuCl₄ is structurally identical to its bromine analog. Partial Br/Cl substitution in (Hpip)₂CuBr₄(1-x)Cl₄x takes place far from the Cu²⁺ sites that carry the \( S = 1/2 \) moments needed for the magnetic properties. It will thus not affect the general properties of the system and its ladder-like structure. It is thus much less violent than other forms of magnetic disorder, such as the replacement of the Cu²⁺ ions by Zn²⁺ (\( S = 0 \)), which fully removes a
Figure 2. Complete Br/Cl substitution series (Hpip)$_2$CuBr$_{4(1-x)}$Cl$_{4x}$, $x = 0$ (left, black) and $x = 1$ (right, yellow).

Figure 3. Schematic temperature–magnetic field phase diagram for the two compounds (Hpip)$_2$CuBr$_4$ and (Hpip)$_2$CuCl$_4$. The coherence scale (see the text) of the order of the exchange $J = J_1$ along the legs of the ladder is shown. Below this coherence scale the system is described by a TLL. If there is a weak interladder residual coupling $J'$, the spin will ultimately show three-dimensional antiferromagnetic planar order (3D).

3. Specific heat measurements

3.1. The theoretical methodology

In order to make predictions that can be compared with experiments, we need to compute e.g. magnetization and specific heat for the Hamiltonian (1) and the coupling constants extracted from the magnetization. This independent check provides a very stringent test for the theoretical description. For the (Hpip)$_2$CuBr$_4$ compound this program has been successfully carried out [17].

We present in this paper a similar calculation of the specific heat for the chloride compound (Hpip)$_2$CuCl$_4$.
3.2. Results and comparisons

Like for (Hpip)_2CuBr_4, we determined for (Hpip)_2CuCl_4 the exchange parameters from the magnetization curves (J_⊥ = 3.42 K, J_∥ = 1.34 K, g = 2.06). They compare favorably with other values found in the literature (table 1, [30]). The two values of J_⊥ and J_∥ are significantly smaller than the ones for (Hpip)_2CuBr_4, confirming the important effects of the Cl substitution on the exchange paths. Note that despite the change of the coupling constants, the general structure of the phase diagram shown in figure 3 is still unchanged, and in particular the lower critical field h_{c1} is still positive, at variance with what happens in similar compounds [29].

The specific heat computed with the fixed exchange parameters is given below. Such calculations when compared with experiments can serve as a benchmark for confirming the Heisenberg form of the Hamiltonian for (Hpip)_2CuCl_4. We show first the high field results h > h_{c2} in figure 4. The presence of a gap is clearly visible in the higher field curves. As the magnetic field moves down, the gap reduces as one approaches h_{c2}, which signals the entrance into the gapless TLL regime.

We show the data within the TLL phase in figure 5 (for a comparison with the (Hpip)_2CuBr_4 findings, see e.g. [17] and figure 6 of [19]). The gap has closed and the low energy part of the specific heat is now linear, as can be expected in a TLL. The slope of the specific heat is directly connected to the speed u of the spin excitation in the TLL by C_m(T) ∝ T/u [2]. Note that a peak structure is shown by the data (most visible for the 3.25 T curve). This peak signals the coherence scale of the TLL (which can also be computed independently from the exchange constants and the filling of the band). It is less visible than for (Hpip)_2CuBr_4 because it is more masked by the contributions to the specific heat coming from the higher triplets in the spectrum.

Finally we show the low field computed data in figure 6. As for the high field data, the gap is again clearly visible and goes down as the field approaches h_{c1}. The specific heat of (Hpip)_2CuCl_4 was measured using a purpose-built calorimeter at the Helmholtz Center Berlin for single crystals with a mass of 3.73 mg between 0.3 and 10 K using both quasi-adiabatic and relaxation techniques. We show raw data for a small magnetic field H = 0.5 T applied parallel to the crystallographic a-axis in figure 7. The lattice contributes to the specific heat at low temperatures, but its signal is distinct from a clear maximum originating from magnetic excitations. The lattice contribution can be subtracted following a procedure that was applied successfully to (Hpip)_2CuBr_4 [17].
The agreement, shown in figure 7, between theory and experiment is remarkable. It thus confirms, like for the parent bromide compound, that no major term is missing from the Hamiltonian (1). It also confirms that the values of the exchange parameters that were determined by an independent method from the critical fields \( h_{c1} \) and \( h_{c2} \) are indeed accurate and more precise than in previous studies [30].

4. Discussion and conclusion

We have presented in this paper a determination of the Hamiltonian and of the exchange parameters that describe the compound \((\text{Hpip})_2\text{CuCl}_4\). The comparison between the measured specific heat of this compound and the calculations, using density matrix renormalization group calculations, show a remarkable agreement between theory and experiment vindicates the form of the Heisenberg Hamiltonian and values of the exchange parameters used.

Figure 7. Comparison between the measured and computed specific heat for \((\text{Hpip})_2\text{CuCl}_4\) at \( H = 0.5 \) T. Lines denote the spin (DMRG with 60 rungs \((2 \times 60 \text{ sites})\) and lattice contributions. The remarkable agreement between theory and experiment vindicates the form of the Heisenberg Hamiltonian and values of the exchange parameters used.

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