Numerical Study of the Kitaev-Heisenberg chain as a spin model of the K-intercalated RuCl$_3$

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Abstract. We study the one-dimensional Kitaev-Heisenberg model as a possible realization of magnetic degrees of freedom of the K-intercalated honeycomb-lattice ruthenium trichloride $\alpha$-RuCl$_3$, denoted as K$_{0.5}$RuCl$_{m}$. First, we discuss the possible charge ordering patterns in K$_{0.5}$RuCl$_{m}$, where half of the $j = \frac{1}{2}$ spins are replaced by nonmagnetic ions in the honeycomb layer. The phase diagram is obtained as a function of nearest-, second-, and third-neighbor repulsions in the atomic limit. We find a strong competition between possible three charge ordering patterns when the repulsions are comparable. We further calculate the spin-spin correlation functions for the 1D Kitaev-Heisenberg model, which is one of the ordering patterns, and consider the effect of the Kitaev term by comparing the results to those for the antiferromagnetic SU(2) Heisenberg chain.

1. Introduction

Since the Kitaev model was proposed in 2006 [1], a quantum spin liquid (QSL) state induced by the Kitaev-type bond-dependent interactions is one of the most active fields in the condensed matter physics. In recent years, because of the emergence of candidate materials and the interest in the QSL realization, there has been a growing number of researches on the Kitaev model, at first, and on the Kitaev-Heisenberg (KH) after. Adding the Heisenberg-type interaction to the initial, exactly integrable, model is necessary and inevitable for any realistic description. However, the interplay between the Kitaev and Heisenberg interactions is always a nontrivial problem to solve. Nevertheless, the vast majority of these studies has been focused on two-dimensional lattices, while research on the KH chain is lacking.

The first candidates for the Kitaev materials are 5$d^5$ honeycomb iridates $A_2$IrO$_3$ ($A=$Na or Li) [2, 3]. These materials are referred to as spin-orbit assisted Mott insulators and the magnetic degrees of freedom can be well described within an effective $j = \frac{1}{2}$ pseudospin model. Due to the existence of two Ir-O-Ir exchange paths with a $\sim 90^\circ$ bond angle in the edge-sharing IrO$_6$ octahedra, a substantial anisotropic Kitaev-type coupling stems from the particular superexchange interactions between the $j = \frac{1}{2}$ pseudospins in addition to the isotropic Heisenberg coupling. It is known that the Kitaev QSL is most likely preempted by a certain level of the Heisenberg interactions. In fact, the above iridates exhibit long-range magnetic order at low temperatures [4, 5, 6, 7], possibly assisted by off-diagonal and/or long-range interactions.

At present, one of the most promising materials close to the Kitaev QSL is the 4$d^{5}$ ruthenium trichloride $\alpha$-RuCl$_3$ in its honeycomb crystal phase. Neutron and Raman scattering studies gave...
In the present context is the chain-type pattern. Very recently, the temperature-dependent electron energy loss spectroscopy (EELS) measurement was performed in K-intercalated RuCl$_3$, denoted as K$_{0.5}$RuCl$_m$, for a fully K-doped sample [17]. The intercalated K$^+$ ions may provide charge carriers, and however, a sharp peak (gap) was observed at $\sim 0.9$ eV, indicating an insulating feature of K$_{0.5}$RuCl$_m$, instead of charge gap $E_g = 1.1$–$1.2$ eV for the undoped $\alpha$-RuCl$_3$ [18, 19, 20]. This has been interpreted as half of the $j = \frac{1}{2}$ pseudospins being replaced by nonmagnetic $d^6$ ions whereas we should see a pseudogap behavior in the EELS spectrum if the intercalated K$^+$ ions are randomly distributed. Therefore, this insulating state can be envisaged as a formation of superlattice by charge disproportionation (charge ordering). Different possible charge ordering patterns are shown in Fig. 1. Of the particular interest in the present context is the chain-type pattern.

In this paper, we consider the charge ordering patterns stabilized by the intersite repulsions and study the 1D KH model as a possible minimal spin model to describe the magnetic properties of the K-intercalated RuCl$_3$. Using the Lanczos exact diagonalization method, we calculate real-space spin-spin correlation functions. The effect of the Kitaev term is discussed by seeing a deviation from the isotropic antiferromagnetic Heisenberg chain. This paper is organized as follows. In Sec. 2, we discuss the possible charge ordering patterns in the classical limit and obtain the phase diagram. We also define the 1D KH chain as a spin model for the chain-type ordering. In Sec. 3 we show the results of spin-spin correlation functions and discuss the effect of the Kitaev term on the antiferromagnetic Heisenberg chain. Section 4 contains summary and conclusions.

2. Model

2.1. Charge ordering pattern in $K_{0.5}RuCl_m$

The insulating feature of $K_{0.5}RuCl_m$ with a sharp peak at $\sim 0.5$ eV in the EELS spectrum may be explained by charge ordering associated with a superlattice formation rather than a random distribution of K$^+$ ions. The superlattice structures are simply constructed by replacing half of the $j_{\text{eff}} = 1/2$ pseudospins with nonmagnetic sites in the original hexagonal cluster. Practically, there are three possibilities of the charge ordering pattern: triangle-, dimer-, and chain-type charge orderings. So far, no crystal structural data of K$_{0.5}$RuCl$_m$ has been available and the detailed calculations are difficult. Thus, we simply consider the charge distribution to minimize the Coulomb repulsions, which are typically of larger order of magnitude than the magnetic interactions. The nearest- ($V_1$), second- ($V_2$), and third-neighbor ($V_3$) repulsions are taken into account (see Figure 1). In the atomic limit (hopping $t = 0$), the energy per particle is $E_t = 3V_2$, $E_d = \frac{1}{2}V_1 + V_2 + V_3$, and $E_c = V_1 + V_2$ for the triangle-, dimer-, and chain-type charge orderings, respectively. For a given set of $V$’s, the dominant configuration is determined as one having the lowest energy. In Figure 1, the obtained phase diagram is shown as a function of $v_1 = V_1/V_2$ and $v_2 = V_3/V_2$. We can see that the three ordering patterns strongly compete when the repulsions $V$’s are comparable. We note that the chain-type ordering always gives the lowest magnetic energy. To decide the detailed charge distribution, experimental observation and/or microscopic analysis including structural distortion by the K-intercalation are indeed required in future.

Since each charge has a $j_{\text{eff}} = 1/2$ pseudospin, the ordered charges are expected to interact magnetically. Recently, for exfoliated $\alpha$-RuCl$_3$, a first-order structural phase transition at $\sim 150$ K between low-temperature C2/m and high-temperature P3$_1$ structures was reported. [21] At
the high temperature an emergence of charge ordering originating from anisotropy in the charge distribution along Ru-Cl-Ru hopping pathways was observed. It suggests a strong tendency to the bond-directional anisotropy along the Ru-Ru axes. In this sense a realization of the chain-type charge ordering may be most likely. Besides, if we assume a magnetic interaction between the nearest-neighbor sites, only the chain-type configuration derives a nontrivial spin model. The triangle-type ordering provides isolated spins and the dimer-type ordering isolated singlet/triplet pairs.

2.2. Hamiltonian
It is generally believed that the magnetic properties of undoped α-RuCl$_3$ is well described by the KH model on a honeycomb lattice. If we assume the chain-type charge ordering, the zigzag chains are well separated. The each chain is considered to be a 1D KH model, which is equivalent to a system obtained by removing the $z$-bonds from the honeycomb-lattice KH model. The Hamiltonian of 1D KH model is written as

$$H = K\sum_{i=1}^{L/2}(S_{2i-1}^x S_{2i}^x + S_{2i}^y S_{2i+1}^y) + J\sum_{i=1}^{L}S_i^z S_{i+1}^z$$

for a system with $L$ sites, where $\vec{S}_i = (S_i^x, S_i^y, S_i^z)$ is a spin-1/2 operator at site $i$. The Kitaev and Heisenberg exchange couplings are denoted as $K$ and $J$, respectively. The system has two kinds of neighboring links and they appear alternately along the chain. Thus, the structural unit cell contains two lattice sites. Hereafter, we call the links $(2i-1, 2i)$ and $(i, 2i+1)$ as “$x$-link” and “$y$-link”, respectively. By rewriting Eq. (1) to

$$H = \frac{2J + K}{4}\sum_{i=1}^{L}(S_i^+ S_{i+1}^- + S_i^- S_{i+1}^+ ) + J\sum_{i=1}^{L}S_i^z S_{i+1}^z + \frac{K}{4}\sum_{i=1}^{L}(-1)^i(S_i^+ S_{i+1}^- + S_i^- S_{i+1}^+ )$$

$$= H_{ex} + H_{Ising} + H_{dsf},$$

we can easily notice that a XXZ Heisenberg chain containing exchange ($H_{ex}$) and Ising ($H_{Ising}$) terms is disturbed by sign-alternating double-spin-flip ($H_{dsf}$) fluctuations.
3. Numerical results
To investigate the effect of the Kitaev term, we calculated the distance dependence of spin-spin correlation functions using the Lanczos exact diagonalization method for a periodic chain with 24 sites. The results for $K = 0, 1,$ and $-0.57$ are plotted as a function of distance $r$ in Figure 2, where the Heisenberg interaction is fixed at $J = 1$. Therefore, we can see a deviation from the antiferromagnetic SU(2) Heisenberg chain ($K = 0$). Interestingly, the total spin-spin correlation function $\langle \vec{S}_0 \cdot \vec{S}_r \rangle$ is almost unchanged. The positive (negative) value of $K$ increases (decreases) quantum fluctuations on the $z$-axis and decreases (increases) along the xy-plane. As a result, their effects roughly cancel each other in $\langle \vec{S}_0 \cdot \vec{S}_r \rangle$. Whereas, the $z$-component $\langle S^z_0 S^z_r \rangle$ is strongly affected by the Kitaev term. Especially, the staggered oscillation is enhanced in the case of negative $K$ value, although the double-spin-flip term is naively expected to weaken it. This means that the Ising term is still dominant in the region $-0.57 \leq K < 0$ with $J = 1$ due to a decrease of the exchange term, and the effect of double-spin-flip term seems not to emerge explicitly there. The other correlations $\langle S^x_0 S^x_r \rangle$ and $\langle S^y_0 S^y_r \rangle$ are oppositely-affected from $\langle S^z_0 S^z_r \rangle$.

4. Summary
It is widely believed that the magnetic properties of $\alpha$-RuCl$_3$ are well described by the honeycomb-lattice $S = \frac{1}{2}$ KH model. First, we studied possible charge ordering patterns in the K-intercalated material K$_{0.5}$RuCl$_{3m}$, where half of the $j = \frac{1}{2}$ spins are replaced by nonmagnetic ions in the honeycomb layer. The phase diagram was obtained as a function of nearest-, second-, and third-neighbor Coulomb repulsions in the atomic limit. We found a strong competition between possible three charge ordering patterns, namely, triangle-, dimer-, chain-type, when the repulsions $V$’s are comparable. Next, using the Lanczos exact diagonalization method we calculated the spin-spin correlation functions $\langle \vec{S}_0 \cdot \vec{S}_r \rangle$, $\langle S^x_0 S^x_r \rangle$, $\langle S^y_0 S^y_r \rangle$, $\langle S^z_0 S^z_r \rangle$ for the 1D KH model, which is derived for the chain-type ordering pattern. Finally, we discussed the effect of the Kitaev term by comparing the correlations to those of the antiferromagnetic SU(2) Heisenberg chain.

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