Project Title:

Numerical study on Kagome-lattice frustrated quantum spin liquids by the iDMRG method

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1. A long-range magnetic order and the associated spontaneous symmetry breaking are commonly observed in most of magnetic materials at low temperatures. In fact, a magnetic order can be suppressed by quantum spin fluctuations and a geometrical frustration. This issue of quantum spin liquids and valence bond solids has long been one of the central issues in condensed matter physics. In particular, spin-1/2 kagome antiferromagnets have been studied as a prototypical model for Mott-insulating frustrated magnets showing a quantum spin liquid, though its nature remains controversial. We have launched the current project to resolve this issue by means of infinite-size density-matrix renormalization group (iDMRG) calculations. By the last fiscal year, we have successfully implemented the methodology for computing the ground state as well as diffuse magnetic neutron-scattering spectra for comparing a theory with experiments on relevant materials. The calculations of these excitation spectra are indispensable for identifying the model parameters.

   To demonstrate the code for these excitation spectra, we have focused on an explanation of an unusual ferroelectric, nonmagnetic and thus spin-gapped ground state of coupled frustrated spin-1/2 chain compound $Rb_2Cu_2Mo_3O_{12}$. Experimental results on the magnetic susceptibility and the inelastic neutron scattering spectra. Electron spin resonance spectra on this compound have also been numerically computed in a related project Q17210. The neutron scattering spectra of powder samples of $Rb_2Cu_2Mo_3O_{12}$ contain three distinct spin excitation levels at 2.0 meV, 0.38 meV and 0.6 meV at the wavevector $Q = 1/4$ r.l.u. along the chain direction (the crystallographic b axis). In the last fiscal year, we reported tentative calculations of the three excited levels in Q16308. This year, we have aimed at fixing such theoretical model parameters that reproduce overall experimental results.

2. The ground state and several low-energy excited states of the model explained below have been computed by (i) the numerical exact-diagonalization method and (ii) the real-space parallelized DMRG method with a four-spin unit for the matrix-product state whose matrix dimension is taken up to 800 to ensure the convergence. We have spent 1.13M hours and 65k hours of CPU time on the MPC and ACSG systems, respectively, by February 15, 2018.

   We have taken as a minimal model to describe magnetic properties and low-energy spectra of $Rb_2Cu_2Mo_3O_{12}$, a frustrated J_1-J_2 spin-1/2 two-leg ladder model with Dzyaloshinskii-Moriya (DM) interactions, namely $H = H_{\mathrm{SU(2)}} + H_{\mathrm{DM}}$. Here, $H_{\mathrm{SU(2)}} = \sum_{\ell}\left[\sum_{\sigma=\pm}\sum_{j=1,2} J_j \boldsymbol{s}_{\sigma,\ell}\cdot\boldsymbol{s}_{\sigma,\ell+1} + J'\boldsymbol{s}_{+,\ell}\cdot\boldsymbol{s}_{-,\ell}\right]$ is the SU(2)-symmetric exchange interaction part with the ferromagnetic first-neighbor coupling

$J_1 < 0$, the antiferromagnetic second-neighbor coupling $J_2 > 0$, and the antiferromagnetic interchain rung coupling $J' > 0$, while $H_{\mathrm{DM}} = \sum_\ell \sum_{\sigma=\pm} \sigma \left[(-1)^\ell \boldsymbol{D}_{\mathrm{s}} \cdot \boldsymbol{s}_{\sigma,\ell} \times \boldsymbol{s}_{\sigma,\ell+1} + \boldsymbol{D}_{\mathrm{u}} \cdot \boldsymbol{s}_{\sigma,\ell} \times \boldsymbol{s}_{\sigma,\ell+1}\right]$ is the DM interaction part $\boldsymbol{D}_{\mathrm{s}} \perp \boldsymbol{D}_{\mathrm{u}}$ in the z and x direction, respectively.

3. The maximum peak position q_0 in the static spin structure factor $s_q^x = \sum_{\ell=1}^{N} \langle s_{+,0}^x s_{+,\ell}^x \rangle e^{-iq\ell/N}$ for $H_{\mathrm{SU(2)}}$ has been computed by the iDMRG method (Fig.1). The region of $q_0 = 1/4$ r.l.u observed in experiments is located at a boundary between blue area and green area. The temperature dependence of the magnetic susceptibility has been calculated by the exact-diagonalization method on a 16-site cluster. A choice of $J_1/J_2 = -3.24$ and $J'/J_2 = 0.583$ reproduce both the period of short-range spin correlations and the temperature dependence of the magnetic susceptibility (Fig.2).

Next, to explain three low-energy spin excitation levels of 0.2 meV, 0.38 meV and 0.6 meV as well as the magnetic susceptibility modified by the DM interactions, we adjust parameters J_2, $D_{\mathrm{u}}/|J_1|$, $D_{\mathrm{s}}/|J_1|$ while J_1/J_2 and J'/J_2 are fixed. The best fits are shown in Fig.3(a) and 3(b) for the exact-diagonalization and 16-site cluster calculations, respectively. (See also Fig.2(b) for the magnetic susceptibility.)

Final results on the powder-averaged neutron-scattering spectra (Fig.4) successfully reproduce the experimental findings.

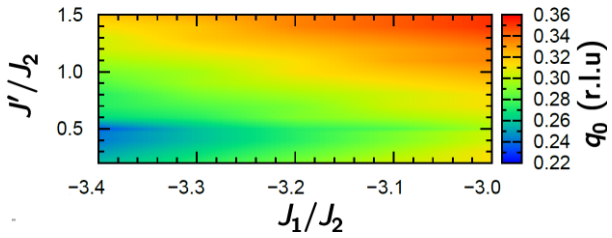

Fig.1 Maximum peak position q_0 in the Fourier component S_q^x for $H_{\mathrm{SU(2)}}$.

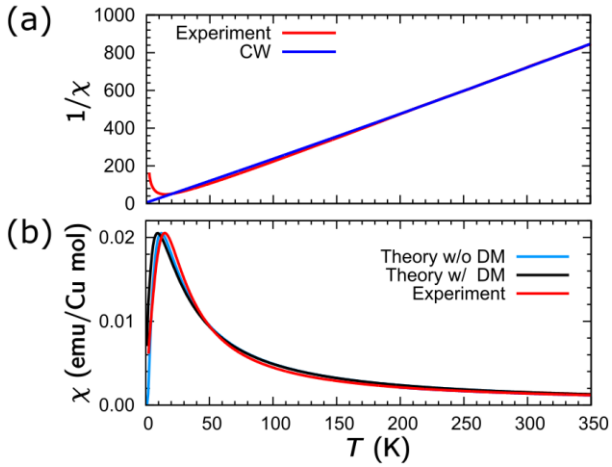

Fig.2 (a) Curie-Weiss fitting of $\chi = \left(\frac{1}{2} g \mu_{\mathrm{B}}\right)^2 / (T - \Theta) + \chi_0$ with $g = 2.16$ and the Bohr magneton μ_{B} for the magnetic susceptibility χ of $B = 0$ in the scale of $1/\chi$, where the best fit has been obtained in a temperature range $T =$ [200,350] K for $\Theta = -2.3$ K and $\chi_0 = -6.1 \times 10^{-5}$ (emu/Cu mol) for the sum of the van Vleck and diamagnetic susceptibilities. (b) Temperature dependence of χ. It has been calculated by the numerical diagonalization of the 16-site cluster. The blue and black curves represent the numerical results for $H_{\mathrm{SU(2)}}$ with $J_2 = 34.4$ K and for $H_{\mathrm{SU(2)}} + H_{\mathrm{DM}}$ with $J_2 =$ 35.1 K, $D_{\mathrm{u}}/|J_1| = 0.215$, and $D_{\mathrm{s}}/|J_1| = 0.39$. In the both cases, $J_1/J_2 = -3.24$ and $J'/J_2 =$ 0.583 are commonly taken.

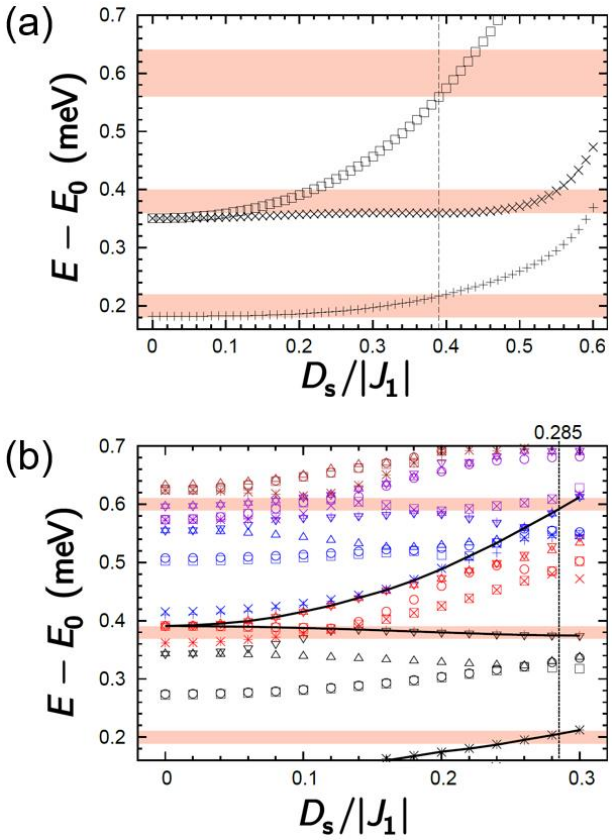

Fig.3: Three lowest-energy levels at $Q_b = 1/4$ r.l.u calculated by the exact diagonalization on a 16-site cluster (a) and the iDMRG (b). The horizontal orange bands point to the three experimentally observed low-energy spin excitation levels 0.20 ± 0.02 meV, 0.38 ± 0.02 meV and 0.60 ± 0.04 meV in (a) and 0.20 ± 0.01 meV, 0.38 ± 0.01 meV and 0.60 ± 0.01 meV in (b). The fitting yields $J_2 = 35.1$ K, $D_u/|J_1| = 0.215$, and $D_s/|J_1| = 0.39$ in (a) and $J_2 = 39$ K, $D_u/|J_1| = 0.182$, $D_s/|J_1| = 0.285$ in (b). Three dashed black curves are guides to the eyes for trajectories of twofold degenerate levels. The choice of $J_1/J_2 = -3.24$ and $J'/J_2 = 0.583$ is commonly taken in the both cases.

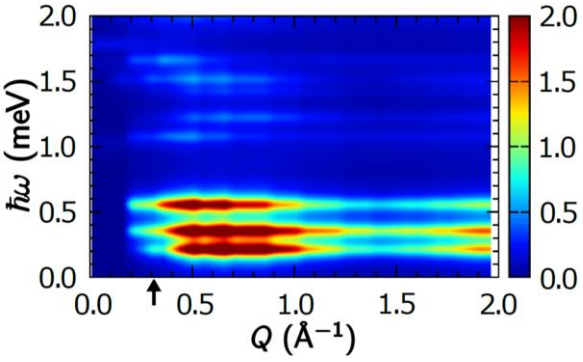

Fig.4: Theoretical low-energy powder-averaged spectra at $B = 0$ and $T = 0$. The position of black arrow corresponds to 1/4 r.l.u.

4. Thus, we conclude the frustrated J_1-J_2 spin-1/2 two-leg ladder model with DM interactions can explain experimental results of the magnetic susceptibility and inelastic neutron scattering spectra of $Rb_2Cu_2Mo_3O_{12}$ in the zero-magnetic field. Our numerical works find the large DM interactions are required to explain the three low-laying energy split from the spin-1 triplet excitation.

5. Although we had a plan to tackle the kagome quantum spin liquid in the spin-1/2 kagome antiferromagnet, it was not possible to achieve this goal in the Quick use, because of the low priority of the jobs. In the next fiscal year, we have a plan to apply the proposal of General use for the calculation to tackle more generic and controversial problems in more generic frustrated magnets.

Fiscal Year 2017 List of Publications Resulting from the Use of the supercomputer

[Oral presentation at an international symposium]

Hiroshi Ueda, Shigeki Onoda, Yasuhiro Yamaguchi , Tsuyoshi Kimura , Daichi Yoshizawa , Masayuki Hagiwara , Masato Hagihara , Minoru Soda , Takatsugu Masuda , Toshio Sakakibara , Keisuke Tomiyasu , Seiko Kawamura , Kenji Nakajima , Ryoichi Kajimoto , Mitsutaka Nakamura , Yasuhiro Inamura , Masashi Hase , Yukio Yasui, “Emergent spin-1 Haldane gap and ferroelectricity in a frustrated spin-1/2 ladder Rb2Cu2Mo3O12”, APS March meeting, V24.00004, March 8, 2018, Los Angeles, USA.