Modeling x-ray photo fluctuation spectroscopy for quantum magnetic systems

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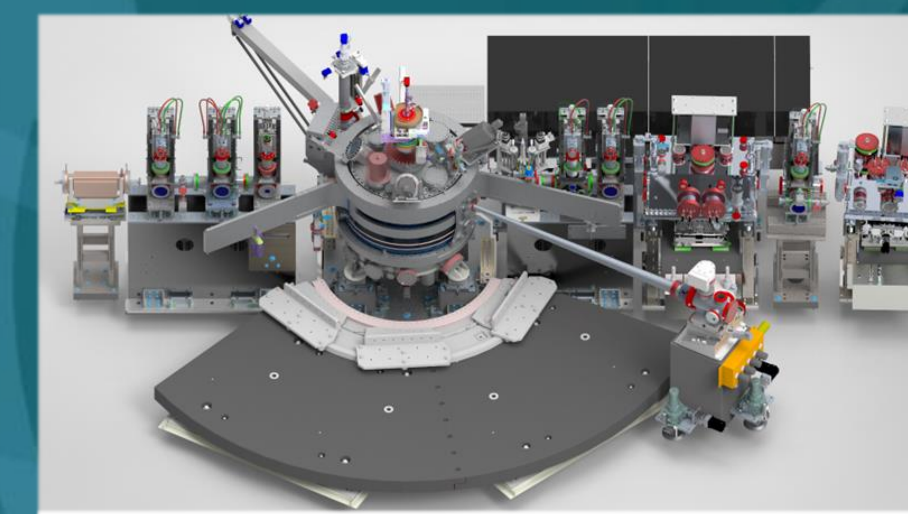
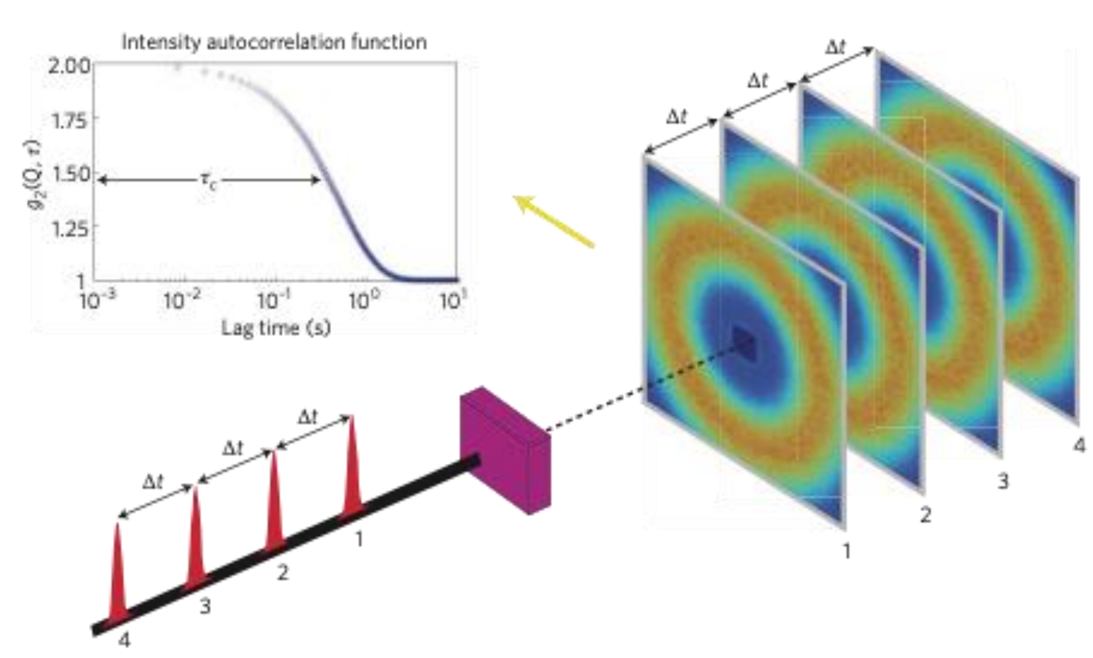


An XPFS chamber! Coming to a national lab near you!

Introduction

The goal of this project was to simulate the intensity autocorrelation function, the "g_2 curve," of x-ray photon correlation spectroscopy (XPCS) measurements for a selection of quantum magnetic models.



A schematic displaying the XPCS technique used to determine a sample's intensity autocorrelation function. [1]

The g_2 curve measures the dynamics of a material based on correlations between intensity spectrums from time-delayed x-ray pulses. g_2 is found as follows:

$$g_2(q,\tau) = \frac{\langle I(q,t) I(q,t+\tau)\rangle_t}{\langle I(q,t)\rangle_t^2},$$

where I(q,t) is the intensity of a RIXS cross-section. Theoretically, it has been shown that the RIXS cross-section measures the dynamical spin structure factor, S(q,t), under certain assumptions. [2] With these conditions met, the g_2 curve can be found as follows:

$$g_2(q,\tau) = \frac{\langle S(q,t) S(q,t+\tau)\rangle_t}{\langle S(q,t)\rangle_t^2}$$

To simulate, S(q,t), it was necessary to find the ground-states of the chosen systems and to simulate the time-evolution of perturbed ground-states. The ground-states were found using the density matrix renormalization group (DMRG) method, while the time evolution was simulated using the time evolution block decimation (TEBD) algorithm. Once S(q,t) is made with these tools, g_2 can be found by calculating the time averages in the above equation.

Models and Materials

Spin-½ Model: $KCuF_3$, Cs_2CoCl_4

$$H = J\sum_{\langle ij\rangle} S_i^x S_j^x + S_i^y S_j^y + \Delta\left(S_i^z S_j^z\right)$$

Spin-1 Model: Ni^{2+}-based chains

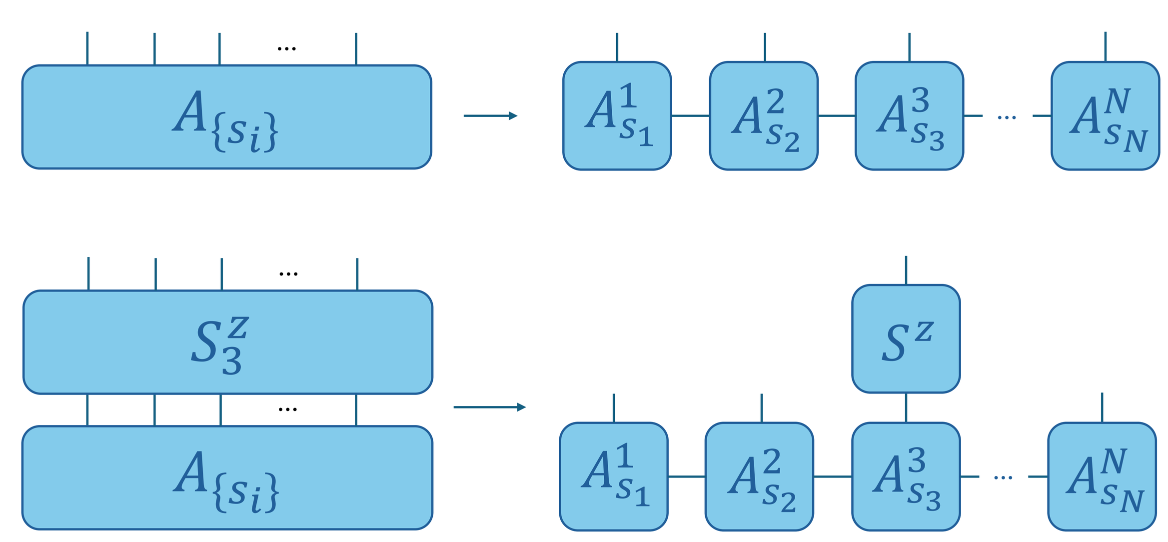
$$H = J\sum_{\langle ij\rangle} S_i \cdot S_j + B_x \sum_i S_i^x + U_{zz}\sum_i (S_i^z)^2$$

Each magnetic model was simulated in a 1D quantum spin-chain system.

Matrix Products and DMRG

To better represent bulk material with a finite simulation, large system sizes are necessary. Finding the ground-state of such a system is no trivial task. The Hilbert space for a typical quantum magnetic system scales exponentially with system size.

Representing quantum states/operators as matrix product states/operators (MPS/O) compresses them into a form that scales linearly with the system size. This form also allows global operators to be applied locally in the Hilbert space of a few sites. Numerical simulations for large systems then become much more computationally feasible.

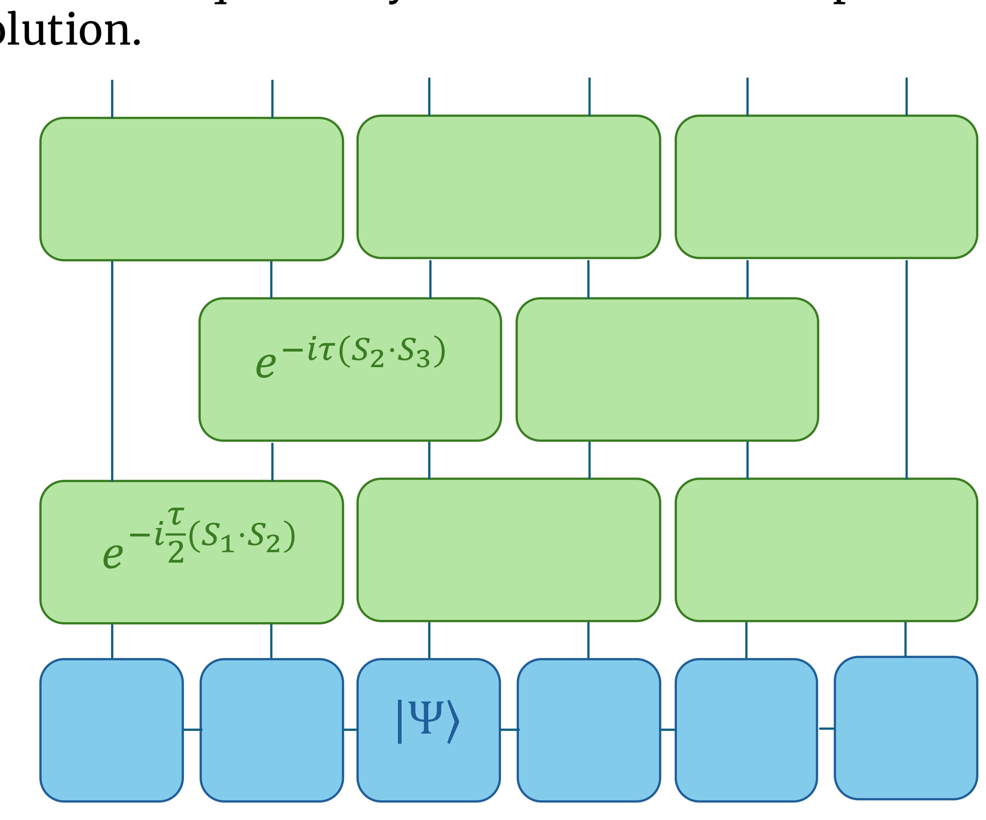


A graphical representation of the advantages of the matrix product representation. (Top) Significant compression of a high-dimensional tensor. (Bottom) Applying global operators as local operators.

The density matrix renormalization group (DMRG) algorithm optimizes an MPS to retrieve the lowest eigenvalue and corresponding eigenstate of a given MPO. In this way, DMRG provides a computationally efficient way to solve the ground-state problem for quantum magnetic systems. [3]

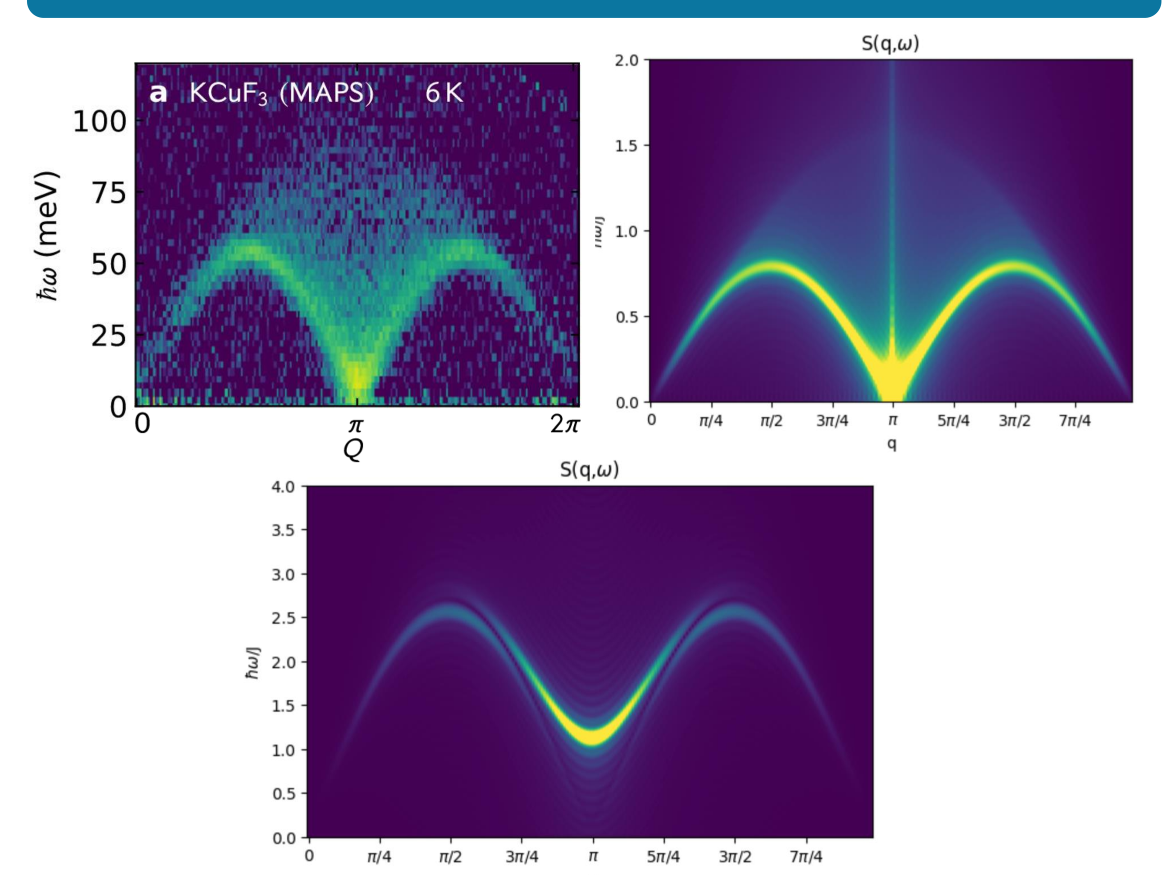
Time Evolution

The TEBD method leverages the matrix product form to efficiently simulate the time evolution of an MPS [4]. With this method, the model's global time evolution propagator is approximated with a series of local propagators that are applied in sets of commuting operators. The error of this approximation scales quadratically with the size of the time step. Thus, short evolutions in time are simulated sequentially to achieve the complete time evolution.



A graphical depiction of how the TEBD method conducts the time evolution of an MPS. This structure is called the Suzuki-Trotter decomposition.

Results

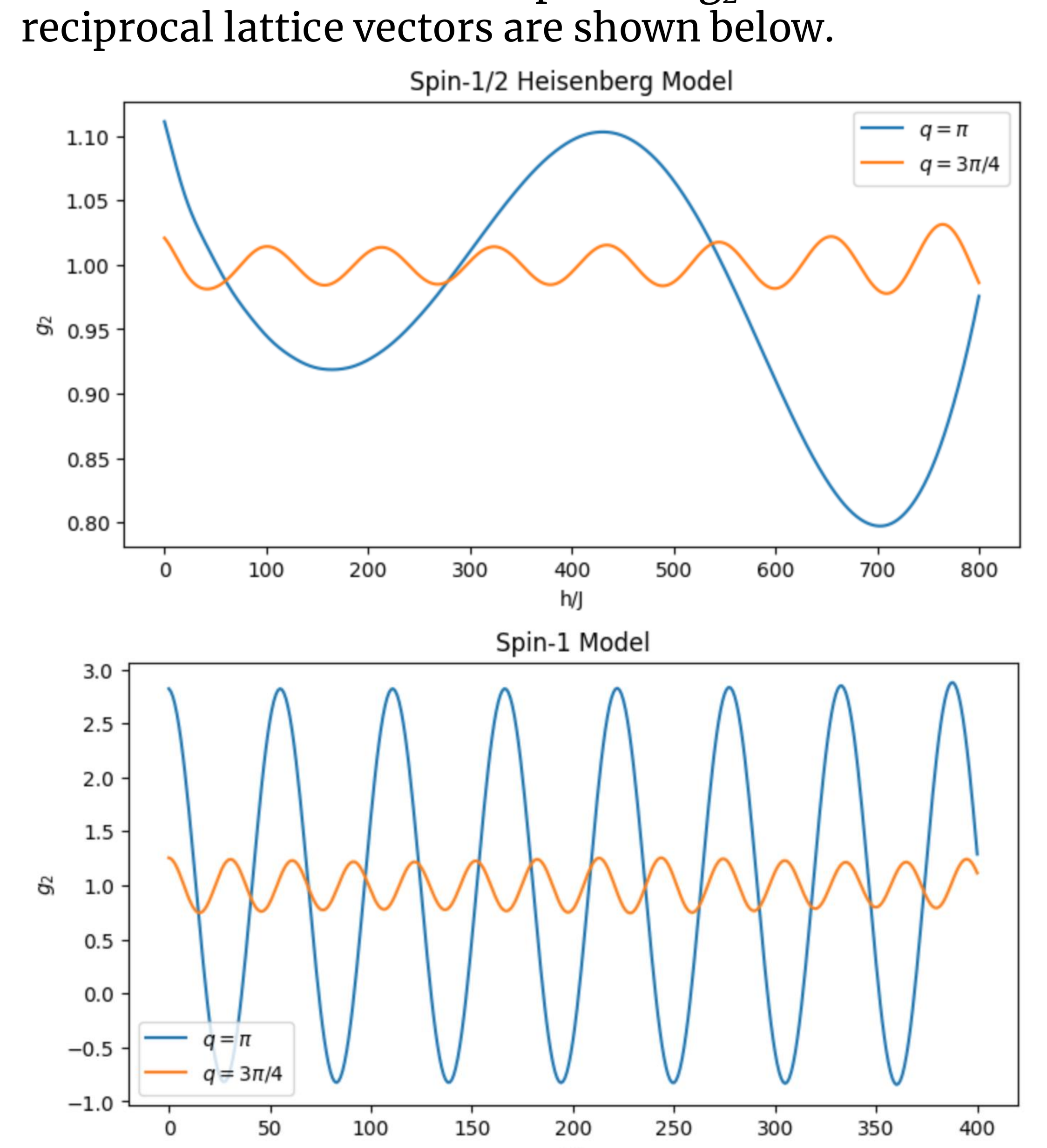


(Top row) A side-by-side comparison of a simulated dynamical spin structure factor measurement with existing neutron scattering results. [5] Color scaling is linear. (Bottom) The same result for the spin-1 model.

To verify the reliability of the simulation, the dynamic spin structure factor result was compared to existing neutron scattering data. The simulation result agreed well with the existing data.

The same result was made for the "Haldane" phase for the spin-1 model. There were clear differences as compared to the spin-1/2 model, most evidenced by the gaps seen in the intensity pattern.

g_2 curves are typically measured at Bragg peaks of interest for a material sample. The g_2 curves for select reciprocal lattice vectors are shown below.



(Top) g_2 curves for the spin-1/2 Heisenberg model, J=1.0, Δ=1.0. (Bottom) g_2 curves for the Haldane phase of the spin-1 model, J=1.0, B_x=0.1, U_{zz}=0.5.

Conclusions and Further Work

The simulation was successfully able to reproduce real neutron scattering data and the g_2 curves found matched expectations. As LCLS prepares to have its very own x-ray photon fluctuation spectroscopy chamber soon, these results pave the way to conduct many experiments at time scales that are unachievable anywhere else in the world!

Acknowledgements

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