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Report on nonlinear magnon processes in antiferromagnets (RU)

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Authors: Hrvoje Vrcan, Johan Mentink

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1. Introduction

1.1. Motivation

The focus of the NIMFEIA project is on making reservoir computing possible by using nonlinear magnonics. So far, many insightful developments have been shown for magnons near the center of the Brillouin zone. However research into nonlinear magnonics at the edge of the Brillouin zone, as relevant to reach higher frequencies, is much less explored.

The treatment of these small and high-frequency magnons is distinct from the classical approach commonly employed in magnonics. First, given the quantum nature of spins, they can no longer be considered classical vectors in the continuum approximation. Then, the wavelengths of very fast magnons get comparable with this lattice discretization where the classical approach stops being valid. For both of these things, a quantum mechanical approach is essential. This is where the field of nonlinear magnonics enters the domain of ultrafast magnetism.

1.2. Nonlinear magnonics meets ultrafast magnetism

A minimal theoretical model necessary for describing the dynamics of spins on the lattice is the Heisenberg model. It is given by the Hamiltonian:

$$H = \sum_{i,\boldsymbol{\delta}} (J_0 + J(t)(\boldsymbol{e} \cdot \boldsymbol{\delta})^2)\boldsymbol{S}_i \cdot \boldsymbol{S}_{i+\boldsymbol{\delta}}.$$

Here, the $\boldsymbol{S}_i = ({S_i}^x, {S_i}^y, {S_i}^z)$ is the vector of quantum spin operators at the site i. The summation is done over the lattice vectors $\boldsymbol{\delta}$ that define crystal bonds between atomic spins.

The constant J_0 is the exchange interaction term that defines the magnetic order of the system. We study antiferromagnetic systems where the exchange constant is positive, favouring antialignment of neighbouring spins. Antiferromagnets have naturally high excitation frequencies [1] and the perturbation of exchange allows us to access the dynamics of the fastest magnons, at the edge of the Brillouin zone.

In [2], it has been summarized how femtosecond laser pulses can excite a magnetic material in a way described by perturbing bonds in a model Hamiltonian, like the Heisenberg model. This excitation is modelled by the $J(t)$ term. The vector $\boldsymbol{e}$ defines the orientation of the electric field of the laser pulse. The maximum effect on a given bond $\boldsymbol{\delta}$ arises when the electric field is polarized along the bond.

To step into the realm of nonlinear magnonics, we must study the dynamics of the Heisenberg model as a function of the shape and the amplitude of the perturbation. There are many prospects beyond the linear response of the system, that can be reached with stronger perturbations. An interesting example of this was already shown by supermagnonic propagation [3]. This shows that magnon-magnon interactions, often considered important only at high perturbation strengths, show up already at relatively weak driving, opening up an exciting possibility for further control with stronger perturbations.

Optical perturbation of the exchange interaction in AFMs leads to the excitation of magnon pairs. The physics of such two-magnon modes is dominated by magnons near the edge of the Brillouin zone and goes beyond the classical description given by the magnetization and the Néel vector, or superpositions of two single-magnon modes [4]. Furthermore, the linearized version of quantum theory which does not include the two-magnon modes, cannot account for the full picture of magnon dynamics (as confirmed by the supermagnonic propagation phenomenon). It will be very interesting to study the effect of nonlinear perturbations on supermagnonic propagation.

2. Method

We can employ several approaches to perform the calculations in the Heisenberg model. The most direct way would be to exactly diagonalize the Hamiltonian, enabling a complete dynamical description derived from the Schrödinger equation. However, this approach is valid only for very small systems. Otherwise, a representational method is required to overcome the exponential growth of the Hilbert space. We chose NQS, a very powerful neural network representation of the quantum many-body wave function. In any case, one must first obtain the ground state of the unperturbed Heisenberg model and then propagate it in time with the selected method's equation of motion.

2.1. Exact diagonalization (ED)

This approach includes using the evolution operator to propagate the system in time. A spin quantum system is represented by a spin configuration, or in other words, written in the product states basis:

$$|\Psi >= |s_1^z > \otimes |s_2^z > \otimes \dots \otimes |s_N^z >= |s_1^z s_2^z \dots s_N^z >.$$

After diagonalizing the unperturbed Hamiltonian in this basis to find the ground state $|\Psi_0 >$, we find the eigenstates $|\phi_m >$ and energies E_m of the full Hamiltonian, and use:

$$|\Psi(t) >= \sum_m < \phi_m|\Psi_0 > e^{-itE_m}|\phi_m >$$

to get the full dynamics. This approach, however, only works for very small systems because the Hilbert space grows as 2^N with the size N, making exact diagonalization unfeasible for larger systems rather quickly.

2.2. Neural Quantum States (NQS)

To overcome this problem with the exact approach, we use a variational representation of the many-body wave function. This approach, pioneered by Carleo and Troyer [5], uses a neural network as a variational ansatz. The network used throughout this research is the Restricted Boltzmann Machine (RBM):

$$\Psi(s) = \prod_j^M 2\cosh\left(b_j + w_{ij} s_i^z\right).$$

Here, the input spin configuration $s = \{s_i^z\}, i = 1, \dots, N$ is passed through a hidden layer with biases b_j and weights w_{ij} to get the probability amplitude $\Psi(s)$ of the given configuration. The ratio $\alpha = M/N$ is set by the user to tune the expressive power of the neural network.

Just like in ED, we first calculate the ground state, equivalent to optimizing the network with gradient descent. To obtain the dynamics, we use the following method.

2.3. Time-dependent variational principle (TDVP)

TDVP is a time-dependent version of the Rayleigh-Ritz variational principle [6]. The cornerstone of this method is the following equation of motion:

$$S_{kk'}\dot{W}_{k'} = -iF_k.$$

Here, the vector $\boldsymbol{W} = (W_1, W_2, \dots, W_M)$ includes all the parameters of the neural network, $\boldsymbol{F} = (F_1, F_2, \dots, F_M)$ is the energy gradient, and the S matrix is the metric of the parameter space.

Since both S and $\boldsymbol{F}$ are functions of $\boldsymbol{W}$, this equation is nonlinear. So, numerical time integration is required to solve it. We use the Heun scheme, which is a second-order update rule:

$$\boldsymbol{W}_{n+1} = \boldsymbol{W}_n + \frac{\mathrm{dt}}{2}\Big(\boldsymbol{f}(\boldsymbol{W}_n) + \boldsymbol{f}\big(\boldsymbol{W}_n + \mathrm{dt}\boldsymbol{f}(\boldsymbol{W}_n)\big)\Big).$$

Here, $\boldsymbol{W}_n$ and $\boldsymbol{W}_{n+1}$ are parameter values at times t_n and $t_{n+1} = t_n + \mathrm{d}t$ respectively, and the update function is formally $\boldsymbol{f} = (f_1, f_2, \dots, f_M) = -iS^{-1}\boldsymbol{F}$.

There's a bit of a caveat here. Usually, due to the singularity of the S matrix, a regularization scheme is used: $S \to S + \varepsilon I$, where the regulator ε is a small number [7]. However, as a mathematical artefact, the regulator is susceptible to noise, as shown in [8]. So, instead of using the regulator, we obtain the update function $\boldsymbol{f}$ by directly solving the TDVP equation of motion without the inversion of the S matrix. In other words, $\boldsymbol{f} = \dot{\boldsymbol{W}}$ that solves the equation $S\dot{\boldsymbol{W}} = -i\boldsymbol{F}$, where a solution is obtained using the least-squares method. This way, we get regularization-free dynamics.

3. Calculations

3.1. Overview

It is known that NQS and TDVP suffer from problems related to the stability of time integration, as indicated by [8]. Here we present the analysis of stability properties depending on the driving strength of the Heisenberg model. We restricted the analysis to a very small system of 2×2 lattice, to keep the exact solutions available and usable as a benchmark for integration methods. We kept the driving in a simple quench form:

$$\mathrm{J(t)} = \begin{cases} 0, & \mathrm{t} < 0, \\ \Delta J_0\,, & \mathrm{t} \geq 0, \end{cases}$$

where the strength of the quench is determined by the Δ parameters. By studying a simple quench scenario, gain insights into the nonlinear dynamics of the two-magnon mode in a minimal model. We are particularly interested in the spin-spin correlation function:

$$C(t) = \langle \boldsymbol{S}_i \cdot \boldsymbol{S}_{i+\boldsymbol{\delta}} \rangle,$$

where $\boldsymbol{\delta}$ indicates the index in the perturbed direction of the lattice. This function was shown to be indicative of the propagation properties of spin waves [3, 4].

3.2. Preliminary exact diagonalization results

We first present the exact diagonalization calculations of the correlation function dynamics. This approach is purely for insight about the correlation dependence on the driving parameter, as a precursor to the NQS approach. We calculated the nearest-neighbour correlation function along the perturbation direction as a function of the driving strength Δ, for insight into the dynamics when we step into the nonlinear regime.

In *Figure 1*, we present some standard behaviour of the correlation function (left). In previous research, the Heisenberg system was driven out of equilibrium by applying a Gaussian pulse with an amplitude of $0.1J_0$, still considered quite a weak driving. We include this value in the quenching scheme, as well as several others in the graph. We can observe a significant influence of the quenching strength on both the amplitude and frequency of the oscillations. Such dependence of the oscillation frequency and amplitude on the quench strength is a hallmark of nonlinear dynamics. A more detailed dependence is shown on the right side of *Figure 1*.

We observe that, by increasing the driving strength in either the positive or the negative direction, we can reach high frequencies and amplitudes of the correlation function, and therefore of propagating spin waves. However, first, we must explore the possibility of reaching these calculations using the NQS method, which scales better to bigger systems.

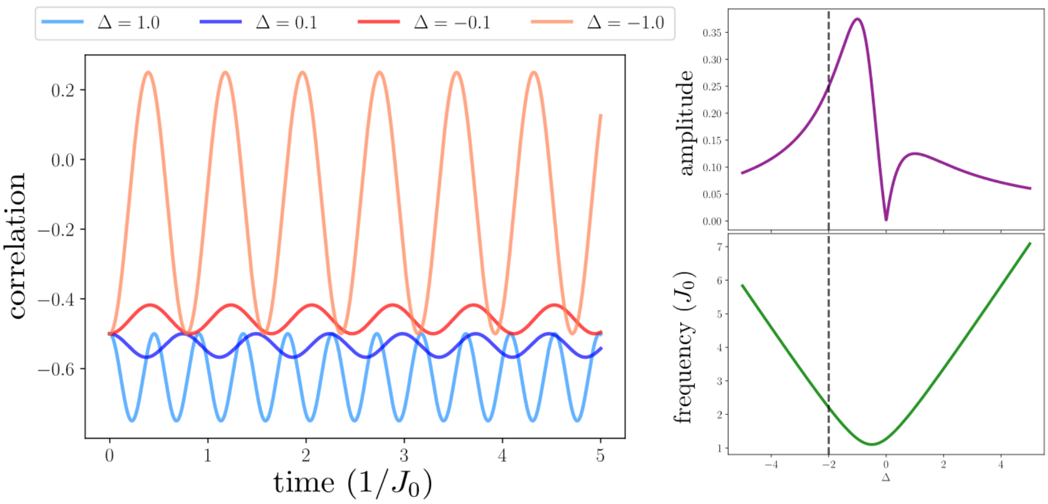

Figure 1. ***Left***: *several examples of the correlation function time dependence for different driving strength* Δ. *The correlation function oscillates in time with a well-defined frequency and constant amplitude. Both can be manipulated by changing the driving parameter.* ***Right***: *amplitude and frequency dependence or correlation function oscillations as functions of the driving parameter. The dashed lines indicate the breakdown scenario, discussed in 3.3.*

3.3. NQS results

Here, we present a similar type of calculation: correlation dynamics as a function of perturbation strength, this time calculated with the NQS representation and the TDVP method for time propagation. In tandem, we present the ED results of the same system, to test the validity of NQS in the regimes of strong driving. This will bring us a step closer to the certainty that we can use this method in the nonlinear regime of dynamics of antiferromagnets.

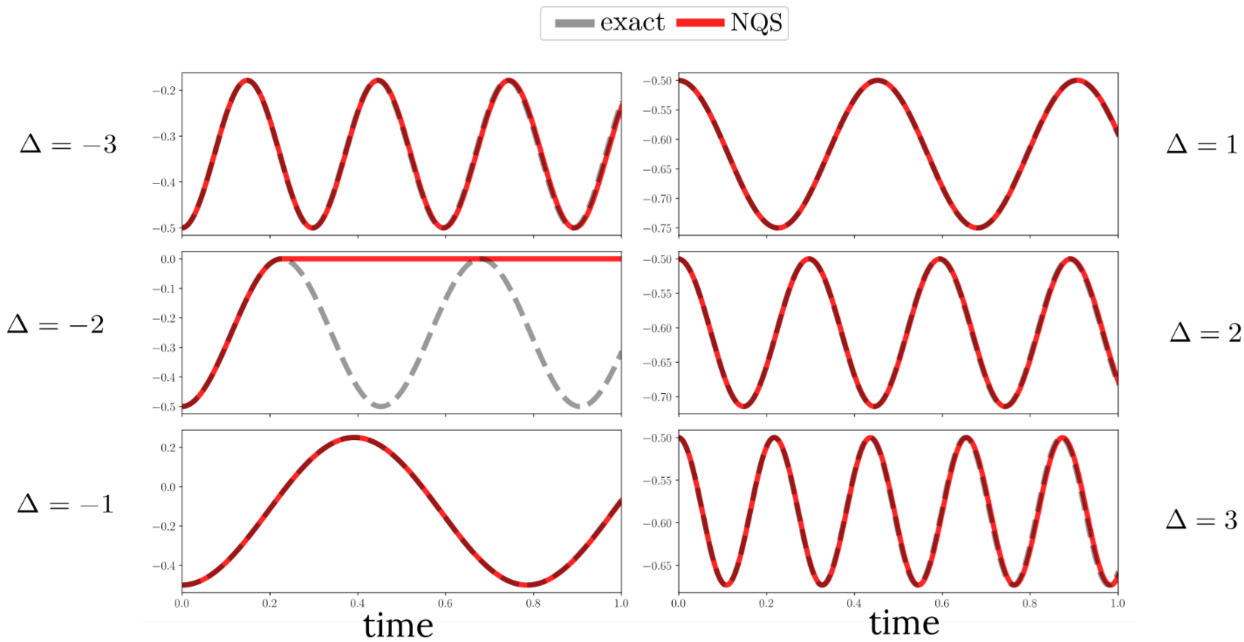

Figure 2. *Correlation dynamics for several values of the quenching parameter, obtained by NQS, as indicated by full red lines. Exact results are presented as a benchmark in grey dashed lines. The results show that NQS faithfully reproduces exact dynamics for all values of the quenching parameter, except for one. At* $\Delta = -2$ *a numerical breakdown occurs.*

In *Figure 2*, the time profile of the correlation function for different values of quenching strength is shown, obtained by both NQS and ED. For the most part, NQS results follow the curves of the exact dynamics very well, with one obvious exception. At $\Delta = -2$, the NQS results deviate from the ED results wildly after reaching the first maximum of the correlation function. In fact, the dynamical behaviour of the system is lost at that point. We dubbed this occurrence the *breakdown regime*.

This numerical instability represents a significant setback in accessing the physics of nonlinear magnonics at the edge of the Brillouin zone. To proceed further, a way around the numerical breakdown that would lead to stable numerical integration is required.

4. Discussion

Looking at these results, we can come up with a conclusion about this preliminary dive into the nonlinear dynamics of magnons at the edge of the Brillouin zone. We've concluded already that we need an exact and quantum tool for probing this physical scenario, as the classical or linear approaches are insufficient.

In this regard, NQS turned out to be a powerful and promising tool, as far as 2D lattice methods go. But, considering the numerical breakdown, there's clearly a very specific physical scenario where this method just doesn't work, even in the absence of stochastic effects that were earlier found to lead to instabilities. We've found this in the quenching scenario with the driving parameter $\Delta = -2$, but there's no telling if this situation repeats for different perturbation shapes and amplitudes. So, to move forward with the investigation of nonlinear magnonics from the quantum perspective, it's imperative that we work towards understanding this numerical breakdown and its influence on the results we obtain.

We already have a lot of insights about the features of the breakdown. For example, we find that this type of numerical instability is unrelated to either the noise, as no stochastic method was applied, or the singularity of the S matrix. We plan to use our findings to move towards a better understanding and possibly solving the breakdown problem, thus opening doors to a more stable and universally applicable method for studying the dynamics of nonlinear magnons at the edge of the Brillouin zone.

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