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EFFECT OF BOUNDARY CONDITIONS ON MAGNETIZATION OF A NANO-SIZE FERROMAGNET

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Abstract. Based on a recently proposed analytic approach we explore the effect of boundary conditions on the magnetization of a finite size Heisenberg ferromagnet. Three types of boundary conditions are discussed on a specific example of the quantum spin S Heisenberg chain. This allows to demonstrate in an explicit form that discreteness of energy spectrum and sensitivity to boundary conditions become essential at the nanometric scale where a finite spin polarization can be achieved at reasonably high temperatures.

Key words: nanomagnets, magnons, magnetization.

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1. INTRODUCTION

In the previous paper [1] we have shown that Bloch description of bulk ferromagnets can be generalized to a crystal of finite size. This results in a formula for magnetization different from the phenomenological expression commonly used to describe finite size deviations from Bloch's $T^{3/2}$ law. Although solving a few inconsistencies of the latter, this generalization assumes periodic boundary conditions (BC) for the magnetic excitations (spin waves). The main justification of this choice of BC is that eigenfunctions of free particles are plane waves for both finite and infinite spaces, establishing a natural continuity relation between the two cases. However, when referring to micro or nano-size magnets a more realistic treatment should take into account the effect of crystal boundaries. The latter was studied via numerical simulations which, however, do not reveal the mechanism of this effect on thermodynamic behavior as expected from an analytic approach. Below we develop such an approach for the case of two relevant types of boundary conditions: fixed (or clamped) and free. We consider a one-dimensional geometry described by the nearest-neighbor spin S Heisenberg isotropic exchange Hamiltonian in the assumption that the system can be described in terms of a free magnon gas and discuss the applicability of this assumption. The choice of $1D$ geometry allows to better emphasize the studied effect. Another important aspect to be kept in mind is that for a, *e.g.* Fe or Co, crystal of a few nanometer size the de-Broglie wavelength of the excitations (*e.g.*, magnons) becomes comparable to the crystal size at temperatures as large as 100 K, implying that full account of their quantum nature, boundary conditions and discreteness of energy spectrum is essential.

2. MAGNON DISPERSION

The single magnon dispersion is given by the following expression [2]

$$E(q) = 2J\sigma(1-\cos q), \tag{1}$$

where q is the wave-vector and the lattice spacing is set to unity, J is the exchange coupling and $\sigma = \langle S_i \rangle$ is the average value of spin per site. The boundary conditions show up in the magnon wave function and momentum. In the ground state $\sigma = S$ and our aim is to calculate the decrease of $\sigma(T)$ with temperature T. As mentioned above, it is important to give a precise meaning to the BC to avoid confusion one may encounter in the literature (*e.g.* "open BC" are used in Ref. [3] in the sense of clamped BC). To this end we write our Hamiltonian defined on a chain of N sites (i.e. of length $N-1$) in an explicit form as

$$H = -J\sum_{i=0}^{N-2}\left(S_i^z S_{i+1}^z + \frac{1}{2}\left(S_i^+ S_{i+1}^- + S_i^- S_{i+1}^+\right)\right) - \left(\mu S_0^z + \nu S_{N-1}^z\right), \tag{2}$$

where μ and ν are external fields acting on the extremities of the chain. The solution of corresponding Schroedinger equation for arbitrary values of parameters has been derived elsewhere [4]- [8]. The BC are controlled by μ and ν so that in the limit $\mu, \nu \to \infty$ we have a chain with *clamped* BC, when no spin flip is allowed at the ends. This is clearly represented by the single magnon amplitude:

$$\begin{gathered} a_C(X) = \sqrt{\frac{2}{N-1}}\sin\left(\frac{\pi m}{N-1}X\right); \\ q_C = \frac{\pi m}{N-1}, m = 1, ..., N-2; \end{gathered} \tag{3}$$

where X runs over the lattice sites. Compare this with the cyclic (*periodic)* chain, when $\mu = \nu = 0$ and $a(N) = a(0)$, with its uniform probability distribution of flipping the spin on a lattice site:

$$a_P(X) = \sqrt{\frac{1}{N}} \exp\left(i\frac{2m\pi X}{N}\right); \quad q_P = \frac{2\pi m}{N}, m = 0, .., N-1. \tag{4}$$

Not surprisingly the two types of states are rather different in energy as well. For an open chain with *free* BC, $\mu = \nu = 0$, we obtain

$$a_F(X) = \sqrt{\frac{2}{N+1}} \cos\left(\frac{\pi m}{N} X + \frac{\pi m}{2N}\right); \quad q_F = \frac{\pi m}{N}, m = 0, .., N-1. \tag{5}$$

In the Fig.1 we compare the three dispersions corresponding to the above BC.

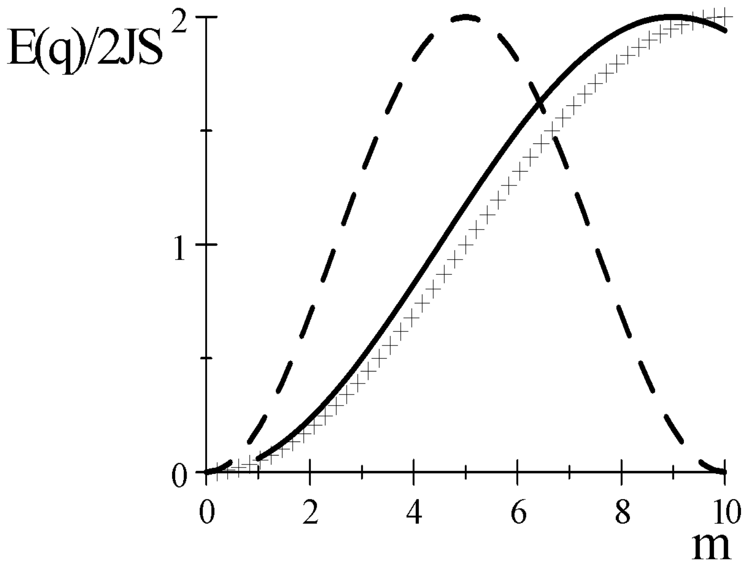

Fig. 1 – Magnon dispersion for periodic (dashed), clamped (continuous) and free (crosses) boundary conditions on a chain of $N = 10$ spins. Only values at integer points m have physical meaning, see also definitions in Eqs. (3)-(5).

Take into account that in the periodic case due to Bloch theorem the descending part of the spectrum should be brought to the first Brillouin zone by translation to negative values of m.

3. MAGNETIZATION *vs*. BOUNDARY CONDITIONS

The spin polarization can now be calculated from the following expression [1]

$$\sigma\left(T\right)=S-\frac{1}{N}\sum_{n=1}^{\infty}\sum_{q\neq\mathbf{0}}\exp-\beta nE\left(q\right), \tag{6}$$

where $\beta=1/k_BT$.Summation over the quantum states determined by the boundary conditions (3)-(5) is based on a generalization of our earlier expression, *e.g.* for the clamped BC we obtain

$$\frac{1}{(N-1)}\sum_{m=0}^{N-1}\exp\left(\beta nJ\cos\left(\frac{\pi m}{N-1}\right)\right)-\frac{\cosh\left(\beta nJ\right)}{(N-1)}=\sum_{k=-\infty}^{\infty}I_{2k(N-1)}\left(\beta nJ\right), \tag{7}$$

where $I_n\left(x\right)$ is the modified Bessel function. Further progress is achieved by realizing that the low temperature expansion is governed by the ratio of temperature and spacing of the energy levels: $N^2/4\pi\beta J\sigma$.It is possible to develop series expansions for both sides of its critical value equal to 1.Below this value we get an exponential series, while above 1 we deal with a power series. In the following we will consider this latter case ($N^2/4\pi\beta J\sigma>1$, $N>>1$), which includes the thermodynamic limit ($N\rightarrow\infty$) as well. It is known (*e.g.* by Mermin-Wagner-Bogolubov theorem) that in this limit σ is strictly zero. Qualitatively, it is due to the dominance of the entropic contribution to the free energy over the energy gained by the full alignment of magnetic moments. However when the number of spin is decreasing so does the entropic term. Therefore one may expect that for a fixed N a certain critical value of $T=T^*$ exists when polarization sets in. This is indeed confirmed by our calculations of the low temperature series generated by the r.h.s. in Eq. (7). Below we show only the main terms of the expansion for the periodic and clamped BC:

$$\begin{aligned}\sigma_P\left(T\right)/S&=1-\frac{1}{NS}\left(\frac{N^2}{4\pi\beta JS}\times1.0472+O\left(\sqrt{\frac{N^2}{4\pi\beta JS}}\right)\right),\\ \sigma_C\left(T\right)/S&=1-\frac{1}{NS}\left(\frac{2N\left(N-2\right)}{4\pi\beta JS}\times1.0472+O\left(\sqrt{\frac{N^2}{4\pi\beta JS}}\right)\right).\end{aligned} \tag{8}$$

Spin polarization corresponding to free BC is somewhat close to that of the clamped BC, as one could anticipate from the similarity of their dispersions shown in the figure.

4. CONCLUSIONS

A few conclusions follow from the expressions in (8). They explicitly show that the leading term of the low temperature expansion for the magnetization of a $1D$

ferromagnet is size dependent (linear in N) and is also linear in temperature. Recall that the standard Bloch theory would produce $T^{1/2}$ and is strictly inapplicable in the $1D$ case. The above equations demonstrate the "mechanism" of the divergence taking place in the thermodynamic limit, that obviously means that no polarization is possible. However, at a finite and still large number of spins N on a chain there exist a finite critical temperature $k_B T^* \sim JS^2/N$ which marks the appearance of spin polarization. Note also the "analogy" of the critical temperature of the $1D$ finite ferromagnet with the mean-field $T_c \sim JS^2$, well suited for bulk $3D$ crystals or for the quasiclassical limit of large spin $S \rightarrow \infty$. Another important conclusion is that periodic boundary conditions underestimate (roughly by a factor of 2) the suppression of magnetization by fluctuations compared to more realistic boundary conditions. This effect is explained by the inhomogeneous distribution over the crystal of the spin flip probability in the latter case. Consequently, spin excitations in the confined regions of the crystal are made easier than in the case of periodic BC. This is also reflected in a softer spectrum of magnon excitations as shown in the figure. Respectively, spin fluctuations are higher for the non-periodic BC, which results in a stronger suppression of magnetization.

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