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Competing magnetic interactions in quantum thin films

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ABSTRACT

In this work we study the quantum spin-1/2 Heisenberg model in two dimensions, with a nearest-neighbor short-range antiferromagnetic exchange (J) and a long-range ferromagnetic dipole–dipole (E_d) coupling. Using the double-time Green's function method within the random phase approximation (RPA) we obtain the magnon dispersion relation as function of frustration parameter δ (δ being the ratio between exchange and dipolar interactions $\delta = J/E_d$). We study the competition between long-range ferromagnetic dipole–dipole interaction and short-range antiferromagnetic exchange in stabilizing the magnetic long-range order in a two-dimensional system. We find that the ferromagnetic order is stable at small k up to critical value of frustration $\delta_c = 0.04375$. For frustration higher than the critical value ($\delta > \delta_c$) our magnetic system is disordered.

1. Introduction

In the past few decades, there has been an increasing interest in understanding the magnetic behavior of systems with low dimensionality, which are characterized by a highly non-linear response to small perturbations [1]. These systems are of interest for fundamental, both theoretical [2–5] and experimental research [6–8], and for technological applications [9]. On a fundamental front, there are interesting questions related to conflicting theoretical and experimental results. Bloch [10] and Mermin–Wagner [11], for example, have established that a two-dimensional (2D) magnetic system cannot display long-range magnetic order at finite temperature (T) when their spins are coupled by isotropic short-range exchange interactions. However, there are few experiments indicating the existence of spontaneous magnetization, at finite T, in ultrathin films [12–19]. This apparent contradiction can in principle be resolved via an *extended* Heisenberg model, where magneto-crystalline anisotropy [20–22] and dipole–dipole interactions [23–25] are simultaneously taken into account.

In the case of the dipole–dipole interaction, it is generally assumed that this interaction is negligible as compared to the exchange interaction [26]. The dipole–dipole interaction is usually long-range and strongly anisotropic in spin space, both properties are important for stabilizing the long-range magnetic order according the Mermin–Wagner [11] theorem. The dipole–dipole interaction can effectively play this role in a 2D system, as shown in [23,24]. Also relevant in this context is the competition between magnetic anisotropy and the exchange interaction. Here we recall, for example, that the magnetic anisotropy in thin ferromagnetic films (based on transition-metal ions) is smaller than the isotropic exchange interaction. In fact, a perusal of experiments reveal that the physical properties of thin films are strongly influenced by magnetic anisotropies in both ferromagnetic and paramagnetic phases. The importance of magnetic anisotropy above the Curie temperature (T_C) is a feature that distinguishes from low- ($\leq 2D$) to three-dimensional systems: in ferromagnetic quasi-2D systems, e.g., $(CN_3NH_3)_2-CuCl_4$, the anisotropy in spin susceptibility was found to survive up to $T/T_C \approx 1.2$ [27]. Moreover, in ultrathin magnetic films, there is clear experimental evidence of an anisotropic magnetic susceptibility also within the high-T paramagnetic phase. This interesting and yet ill-understood behavior was found, for example by Back et al. in epitaxial Fe/W(110) films [28] as well as by Jensen et al. [29] in Co films grown on a vicinal Cu substrate.

Importantly, several experimental techniques have been used to study magnetic excitations in ultrathin films. Brillouin Light Scattering has been employed by Tacchi et al. [2] to investigate the nature of magnetic excitations in few-layers thin films [NiFe/Cu(110) and Fe/Cu(110)], characterized by strong in-plane, uniaxial anisotropy. For temperatures well below T_C, the magnon frequency displays a non-monotonic field dependence for magnetic fields applied along the hard, in-plane direction. However, for fields pointing along the easy direction the magnon frequency

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increases almost linearly with increasing the magnetic field. An interesting aspect here is the fact that the magnetic-excitation anisotropy persists in the paramagnetic phase, i.e., the magnon frequency was found to increase almost linearly with the applied magnetic field, showing different slopes depending on the field direction. These results were confirmed by theoretical calculations using a finite-T Green's function formalism [30], which was developed to study field-driven reorientation transitions in anisotropic monolayers.

Also widely relevant is the attempt to understand the competition between short- and long-range interactions [31,32], in systems with localized spins, in nanoparticle systems [33,34] as well as in novel hybrid systems consisting of a quantum dot and graphene nanodisk embedded in a non-linear photonic crystal [35]. In fact, both the dipole–dipole and exchange interaction are expected to induce slow spin-wave excitation spectrum in real magnetic systems. However, if a small spin anisotropy is present in the system, the spin-wave can lead to significant quantum effects. We recall that ferromagnetic systems governed by a dipole–dipole interaction were studied within the spin-wave formalism by Cohen and Keffer [36], showing considerable zero-point energy deviations of the ground state in *bcc* and *fcc* lattices. Additionally, Sousa and Branco [32] studied the 2D quantum spin-1/2 Heisenberg model with nearest-neighbor antiferromagnetic exchange interactions ($J<0$). They also considered part of the dipole–dipole interaction ($E_d \overrightarrow{S}_i \cdot \overrightarrow{S}_j/r^3$) in their framework. In [32], the dipole–dipole interaction connects each spin to every other spin of the square lattice. Using the Green's function method in the random phase approximation (RPA), they showed that the magnetic order disappear for a certain value of frustration parameter $\delta = J/E_d$.

In this paper we use the Green's function theory to analyze the competition between the magnetic dipole–dipole and exchange interactions in a thin spin-1/2 Heisenberg film, consisting of two single layers on a simple cubic (001) lattice. The remainder of the paper is organized as follows: the model Hamiltonian and theoretical method are described in Section 2. In Section 3, the excitation energy of system is calculated and discussed. Finally, our conclusions are given in Section 4.

2. Model and formalism

In this work we study a two-dimensional spin-1/2 Heisenberg model with short-($J<0$) and long-range interactions which is described by the Hamiltonian

$$\mathcal{H} = \mathcal{H}_{ex} + \mathcal{H}_{dd}. \tag{1}$$

We consider in-plane and inter-plane antiferromagnetic exchange interactions (in H_{ex}) as well as dipole–dipole interaction (H_{dd}) connecting all the spins of the lattice. The first term in Eq. (1) refers to the exchange interaction (short-range) and it is written as

$$\mathcal{H}_{ex} = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j. \tag{2}$$

Here, a spin S is localized on sites of two infinite square lattices parallel to the (z,x) plane, with lattice constant a and distance y between the two planes. Both lattices are in atop geometry, corresponding to a tetragonal structure, and in particular to a (010) simple cubic system for $y=a$ (where a is the lattice parameter). The summation in Eq. (2) runs over all distinct nearest-neighbors spin pairs. The second term in Eq. (1) is the dipole–dipole interaction (long-range), which reads

$$\mathcal{H}_{dd} = \frac{1}{2} g^2 \mu_B^2 \sum_{i \neq j} \sum_j \frac{1}{R_{ij}^3} \left\{ \mathbf{S}_i \cdot \mathbf{S}_j - \frac{3}{R_{ij}^2} (\mathbf{S}_i \cdot \mathbf{R}_{ij})(\mathbf{S}_j \cdot \mathbf{R}_{ij}) \right\}, \tag{3}$$

where g is the Landé factor and μ_B is the Bohr magneton. The summations in Eq. (3) are done over all possible (or distinct) spin pairs in the lattice. In addition, $\mathbf{R}_{ij}$ denotes a vector joining two different lattice sites i and j, which is written as $\mathbf{R}_{ij} = \mathbf{R}_i - \mathbf{R}_j$. Without loss of generality, we choose the magnetization along the z-axis, more precisely in the (z,x) plane, and assume intra- and inter-planar antiferromagnetic order. To gain some understanding of the low-T physics hidden in Eq. (1), we derive below the Tyablikov–Zubarev [37–39] spin double-time Green's function

$$G_{ij}^{+-}(t,t') = -i\theta(t-t') \langle\langle [S_i^+(t); S_j^-(t')] \rangle\rangle, \tag{4}$$

whose Fourier transform with respect to $(t-t')$ is denoted as $\langle\langle S_i^+; S_j^- \rangle\rangle_\omega$. For practical quantum calculations we write the Fourier transform (FT) relative to time in the equation of motion as

$$\omega \langle\langle S_l^+; S_m^- \rangle\rangle_\omega = \frac{1}{2\pi} \langle [S_l^+, S_m^-] \rangle + \langle\langle [S_l^+, \mathcal{H}]; S_m^- \rangle\rangle_\omega. \tag{5}$$

In this equation, the Green's function in the second term on the right-hand side contains a greater number of spin operators than the initial one. So, to reduce the order of the Green's function we apply the random phase approximation (RPA) by introducing factorizations of the form $\langle\langle S_l^z S_j^+; S_m^- \rangle\rangle \cong \langle S_l^z \rangle \langle\langle S_j^+; S_m^- \rangle\rangle$, where $\langle S^z \rangle$ is the statistical average. To simplify our calculations, in the following we assume that all sites of the system are equivalent meaning that $\langle S_l^z \rangle \equiv \sigma$ is independent of the index l ($=1,2$), and therefore it is only a function of T. The first approximation to be made is to neglect Green's functions of the form $\langle\langle S_l^z S_j^z; S_m^- \rangle\rangle_\omega$, $\langle\langle S_j^+ S_l^+; S_m^- \rangle\rangle_\omega$, and $\langle\langle S_l^+ S_j^-; S_m^- \rangle\rangle_\omega$, which allow us to neglect higher order spin fluctuations terms [25]. Indeed, some correlation effects arising from these terms can also be eliminated by a correction to the Weiss ferromagnetic ground state [25]. Therefore, one expects that the resulting angle rotation of local spins would be of the order of the small dipolar anisotropy parameter, i.e., the long-range ferromagnetic dipole–dipole coupling, which we define as $E_d \equiv (g\mu_B)^2/a^3$. These contributions are expected not alter in any significantly way the results for the dispersion relations and, consequently, for the Curie temperature T_C. On the other hand, we must keep all terms quadratic in S^+ and S^- operators, since the guiding line in treating our total Hamiltonian $\mathcal{H}$ (Eq. (1)) is to transform it into a quadratic form representing a collection of harmonic oscillators. Using the above approximations, the equation of motion reads

$$\begin{aligned} \omega \langle\langle S_l^+; S_m^- \rangle\rangle_\omega = {} & 2 \sum_{j \neq l} J_{lj} \left[\langle\langle S_l^z S_j^+; S_m^- \rangle\rangle_\omega - \langle\langle S_j^z S_l^+; S_m^- \rangle\rangle_\omega \right] \\ & - \frac{g^2 \mu_B^2}{2} \sum_{j \neq l} \left\{ \frac{1}{R_{lj}^3} \left(1 - \frac{3Z_{lj}^2}{R_{lj}^2} \right) \left[\langle\langle S_l^z S_j^+; S_m^- \rangle\rangle_\omega \right. \right. \\ & \left. \left. + 2 \langle\langle S_j^z S_l^+; S_m^- \rangle\rangle_\omega \right] - 3 B_{lj}^* \langle\langle S_l^z S_j^-; S_m^- \rangle\rangle_\omega \right\} + \frac{1}{\pi} \langle S_l^z \rangle \delta_{lm}, \end{aligned} \tag{6}$$

where $B_{ij}^* = (R_{ij}^+)^2 / |R_{ij}|^5$ and Z_{ij} is the component of $\mathbf{R}_{ij}$ in the z-direction, with $R_{ij}^+ = R_{ij}^x + iR_{ij}^y$. The application of the RPA represents our second approximation, therefore giving

$$\begin{aligned} \omega G_{lm}^{+-}(\omega) = {} & 2 \sum_{j \neq l} J_{lj} \left[\langle S_l^z \rangle G_{jm}^{+-}(\omega) - \langle S_j^z \rangle G_{lm}^{+-}(\omega) \right] \\ & - \frac{\gamma^2}{2} \sum_{j \neq l} \left\{ \frac{1}{\mathbf{R}_{lj}^3} \left(1 - \frac{3Z_{lj}^2}{\mathbf{R}_{lj}^2} \right) \left[\langle S_l^z \rangle G_{jm}^{+-}(\omega) \right. \right. \end{aligned}$$