Anisotropy effects on the Magnetic Excitations of a Ferromagnetic Monolayer below and above the Curie Temperature

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The field-driven reorientation transition of an anisotropic ferromagnetic monolayer is studied within the context of a finite-temperature Green’s function theory. The equilibrium state and the field dependence of the magnon energy gap $E_0$ are calculated for static magnetic field $H$ applied in plane along an easy or a hard axis. In the latter case, the in-plane reorientation of the magnetization is shown to be continuous at $T = 0$, in agreement with free spin wave theory, and discontinuous at finite temperature $T > 0$, in contrast with the prediction of mean field theory. The discontinuity in the orientation angle creates a jump in the magnon energy gap, and it is the reason why, for $T > 0$, the energy does not go to zero at the reorientation field. Above the Curie temperature $T_C$, the magnon energy gap $E_0(H)$ vanishes for $H = 0$ both in the easy and in the hard case. As $H$ is increased, the gap is found to increase almost linearly with $H$, but with different slopes depending on the field orientation. In particular, the slope is smaller when $H$ is along the hard axis. Such a magnetic anisotropy of the spin wave energies is shown to persist well above $T_C$ ($T \approx 1.2 T_C$).

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I. INTRODUCTION

A great experimental achievement is the ability to grow epitaxially well defined monolayer and sub-monolayer ferromagnetic films. Such films serve as model systems in which to study basic aspects of magnetic ordering in low dimensions as well as to test theories of magnetism. Results gleaned from these studies have wide impact across several areas from critical phenomena to electronic band structure theory. There are also important implications for applications because the issues involved lie also at the heart of problems in interface magnetism. A problem of particular importance is the formation of magnetic anisotropies in thin transition metal magnetic films.

Critical phenomena in low dimensional magnets and the formation of magnetic anisotropies are linked together at a fundamental level. Long range order in two dimensional magnets depends upon the long wavelength behaviour of the Goldstone mode of the spin system. This behaviour is strongly affected by the existence of a zero momentum energy gap. In most real systems, the largest contribution to this gap comes from second order corrections to the exchange energy associated with electronic correlations creating the ordered state. These corrections appear in the form of magnetic anisotropies determined by spatial symmetries of the local atomic environment.

Anisotropy formation is a higher order effect in the sense that the energies involved are much less than the energy associated with the ordering temperature $T_C$. In two dimensional systems, the local fields responsible for magnetic anisotropies are very different from the local fields existing in three dimensional bulk systems, and the same reduction in dimensionality has a profound effect on the critical behaviour of the system. In real magnetic systems accessible to experimental study, the formation of magnetic anisotropies and the critical behaviour of the magnetization are therefore intrinsically linked.

In this paper we show that there are striking features associated with magnetic anisotropies which appear in spectra produced by spin wave excitations in two dimensional ferromagnets. There are very clear and measurable finite temperature effects produced by anisotropies that can be observed below the ordering temperature, and we demonstrate that one can also observe easy and hard directionality in magnetic excitation energies above the ordering temperature.

A few experiments in epitaxial ultrathin ferromagnetic films have explored the nature of the phase transition and the persistence of magnetic anisotropies well above $T_C$. A similar effect was observed in quasi-two-dimensional easy-plane ferromagnets. The role of the energy gap in stabilizing ferromagnetic order in two dimensions is well known in critical phenomena, but less well understood in terms of experiment. Experimentally, the gap can be observed from the spectra of long wavelength spin waves, and can be most clearly seen by taking the magnetic system through a reorientational transition with the application of a static magnetic field along a hard anisotropy direction. Theoretically, the equilibrium configuration can be determined, e.g. by the vanishing of the total static torque acting on spins: for small fields the magnetization orients between the direction of the field and the closest symmetry axis of...
the anisotropy until, at some specific critical magnetic field strength \( H_c \), a reorientation of the magnetization along the field direction occurs. As regards the spin wave excitations away of the equilibrium state, at low temperatures their energy can be easily calculated, using the Holstein-Primakoff transformation from spin operators to boson operators,\(^{6,7,8,9}\). For \( H = 0 \) the spin wave energy is essentially determined by the anisotropy but, as \( H \) increases, the energy barrier to deviate away from the easy axis gradually decreases until, for \( H \) greater than \( H_c \), the field itself constitutes an energy barrier. This results in a minimum value for the energy of a spin wave at \( H_c \). Within free spin wave theory, valid at low temperatures, this minimum is predicted to be exactly zero for zero wavevector.\(^{9}\) It is the aim of the present paper to investigate, at higher temperatures, the field dependence of the magnetization modulus in the same field range. As a consequence, for finite temperature \( 0 < T < T_C \), we find that the two theories give qualitatively different results as regards the order of the field-induced transition: within MFT the reorientation of the magnetization is continuous and the magnon energy gap vanishes at the reorientation field; within GFT the reorientation is discontinuous and the gap does not vanish at \( H_c \). Above \( T_C \), both theories are able to account for anisotropy effects on the energy of the magnetic excitations, but quantitative differences are present. Finally, the conclusions are drawn in Section IV.

II. MEAN FIELD THEORY

We consider a magnetic monolayer with Hamiltonian

\[
\mathcal{H} = \frac{J}{2} \sum_{kl} \mathbf{S}_k \cdot \mathbf{S}_l - K_2 \sum_k (S^Z_k)^2 - g\mu_B H \sum_k \mathbf{S}_k + \frac{1}{2} \frac{(g\mu_B)^2}{a^3} \sum_{k \neq l} \frac{a^3}{r_{kl}^3} \left[ \mathbf{S}_k \cdot \mathbf{S}_l - 3 \frac{\langle \mathbf{S}_k \cdot \mathbf{r}_{kl} \rangle \langle \mathbf{S}_l \cdot \mathbf{r}_{kl} \rangle}{r_{kl}^2} \right] \tag{1}
\]

where \( J > 0 \) is the nearest neighbor ferromagnetic exchange interaction, \( K_2 > 0 \) is a uniaxial single-ion anisotropy favoring the \( Z \) direction in the monolayer plane \((XZ)\), and \( \mathbf{H} = (H^X, 0, H^Z) \) is an external magnetic field applied in plane: see Fig. 1. The last term in Eq. (1) is the magnetic dipole-dipole interaction, favoring the alignment of the magnetization within the film plane; \( \mathbf{r}_{kl} \) denotes a vector joining two different lattice sites and \( a \) is the lattice constant. In the following we will assume a simple quadratic lattice, so that the number of nearest neighbors is 4.

A. Equilibrium condition

The effective field \( H_{e\parallel} \) acting on a spin in the ferromagnetic film is composed of an exchange field \( H_{e\parallel} = \frac{4J\langle S^z \rangle}{g\mu_B} \); a shape demagnetizing field of modulus \( 4\pi g\mu_B \langle S^z \rangle \) favouring the film plane \((Y = 0)\); a uniaxial anisotropy field \( H_K = \frac{2K_2\langle S^Z \rangle}{g\mu_B} \) favouring the crystallographic \( Z \) direction in the film plane; an external field \( H \) applied within the film plane, along the easy \((Z)\) or the hard \((X)\) axis. By \( \langle S^Z \rangle \) and \( \langle S^z \rangle \) we denote, respectively, the thermal average of the spin component referred to the crystallographic reference frame \( X, Y, Z \) and to the local reference frame \( x, y, z \) (see Fig. 1). More precisely, \( z \) is the equilibrium direction of the in-plane magnetization, forming an angle \( \phi \) with the easy in-plane crystallographic direction \( Z \). In the mean field approximation, the thermal average \( \langle S^z \rangle \) referred to the local frame can be calculated self consistently in terms of the classical Langevin function \( \langle S^z \rangle = SL(x) \), where

\[
\mathcal{L}(x) = \coth x - \frac{1}{x} \text{ and } x = \frac{g\mu_B H_{e\parallel} S^t}{k_B T}.
\]

The exchange field is parallel to the magnetization while the shape demagnetizing field does not provide any preference for a particular in-plane direction. The equilibrium value for the angle \( \phi \) that the magnetization forms with the crystallographic easy axis \( Z \) is determined by requiring the vanishing of the static torque \( \mathbf{M} \times (\mathbf{H} + H_K \hat{Z}) \).

For field along the easy axis, \( \mathbf{H} = H^Z \hat{Z} \), there is no competition between the applied field and the anisotropy field, so that \( \phi = 0 \) for any field value. The thermal average of the magnetization is obtained solving \( \langle S^z \rangle = S\mathcal{L}(x) \) with

\[
x_{\text{easy}} = \frac{g\mu_B H \hat{Z} + 2K_2 \langle S^z \rangle + 4J\langle S^z \rangle}{k_B T}.
\]

For field along the hard axis, \( \mathbf{H} = H^X \hat{X} \), the modulus of the anisotropy field is \( H_K = \frac{2K_2\langle S^z \rangle \cos \phi}{g\mu_B} \). In this case, the condition for vanishing static torque is

\[
H^X \cos \phi = \frac{2K_2\langle S^z \rangle}{g\mu_B} \cos \phi \sin \phi \tag{2}
\]
The thermal average is obtained solving the self consistent equation \( \langle S^z \rangle = SL(x) \) with \( x = x_{\text{hard}} = \frac{S}{k_B T} (g\mu_B H^X \sin \phi + 2K_2 \langle S^z \rangle \cos^2 \phi + 4J \langle S^z \rangle) \). As \( H^X \) is increased from zero to the critical field value \( H_c^X = \frac{2K_2 \langle S^z \rangle}{g\mu_B} \), it turns out that \( \phi \) ranges continuously between 0 and \( \pi/2 \) and the magnetization modulus \( M = \langle S^z \rangle \) is independent of the field intensity in the whole field range \( 0 \leq H^X \leq H_c^X \) (see Fig. 2a).

The latter feature can be readily accounted for observing that, upon substituting the equilibrium condition appropriate for \( 0 \leq H^X \leq H_c^X \) (namely \( \phi = \sin^{-1} \left( \frac{g\mu_B H^X}{2K_2 \langle S^z \rangle} \right) \) in the expression for \( x_{\text{hard}} \), one obtains that the magnetization modulus \( M = \langle S^z \rangle = SL(x) \) is determined by the solution of the self consistent equation \( x = \frac{S}{k_B T} (2K_2 + 4J/(\coth x - \frac{\pi}{2})) \), (where the field intensity \( H^X \) does not explicitly appear) in the field range from \( H^X = 0 \) up to \( H^X = H_c^X \). In contrast, for \( H^X > H_c^X \) the magnetization is field dependent since one has \( \phi = \pi/2 \) and \( x = \frac{S}{k_B T} [g\mu_B H^X + 4J \langle S^z \rangle (\coth x - \frac{\pi}{2})] \). Notice that the field dependence of \( M \) for \( H^X > H_c^X \) is not apparent from Fig. 2a since it is very weak for the chosen parameter values.

For zero field, one has \( x = \frac{S}{k_B T} (4J + 2K_2) \langle S^z \rangle \) and the Curie temperature \( T_C \) can be calculated, as usual, by requiring that the initial slope of \( L(x) \) is equal to 1, i.e. to the slope of \( \langle S^z \rangle / S \). One obtains

\[
k_B T_C = \frac{1}{3} S^2 (4J + 2K_2). \tag{3}\]

### B. Resonance frequency

The resonance frequency can now be calculated by solving the linearized Bloch torque equations \( d\mathbf{M}/dt = -\gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} \) where \( \gamma \) is the gyromagnetic ratio. For field along the easy axis, \( \mathbf{H} = H^Z \mathbf{Z} \), the time varying magnetization can be assumed to be of the form \( \mathbf{M} = (\hat{x} m_x + \hat{y} m_y) e^{-i\omega t} + \hat{z} [g\mu_B \langle S^z \rangle] \), while the effective field is

\[
\mathbf{H}_{\text{eff}} = -\hat{y} 4\pi m_y + \hat{z} \left( H^Z + \frac{2K_2 \langle S^z \rangle}{g\mu_B} \right). \tag{4}\]

We note that the effective exchange field was not included in Eq. (4) since it is parallel to the magnetization, thus it does not contribute to the uniform mode frequency. The resonance frequency is then readily found to be

\[
\omega_0^{\text{easy}} = \gamma \left[ H^Z + \frac{2K_2 \langle S^z \rangle}{g\mu_B} \right]^{\frac{1}{2}} \times \left[ H^Z + \frac{2K_2 \langle S^z \rangle}{g\mu_B} + 4\pi g\mu_B \langle S^z \rangle \right]^{\frac{1}{2}}. \tag{5}\]
FIG. 2: Comparison between the results of mean field theory (MFT, left) and Green’s function theory (GFT, right) for the magnetization components (above) and the energy gap (below) versus the intensity $H$ of a magnetic field applied in plane along the hard direction ($X$), for fixed temperature $T/T_C = 0.5$. Figs. 2a,b display the calculated equilibrium angle $\phi$ (thin full line, referred to the right vertical axis), magnetization modulus $M = \langle S^z \rangle$ (dotted line) and magnetization components, $\langle S^z \rangle = M \cos \phi$ and $\langle S^X \rangle = M \sin \phi$ (thick full lines), versus $H$. Figs. 2c,d display the calculated equilibrium angle $\phi$ (thin full line) and energy gap $E_0$ (thick full line) versus $H$. The Hamiltonian parameters used for the calculations are $J = 500$ K, $K_2 = 5$ K, $4\pi \omega f_w = 1$ K, $S = 1$, providing $T_C = 670$ K in mean field theory and $T_C = 502.4$ K in Green’s function theory.

where $\langle S^z \rangle = S L(x)$ and $x = x^{\text{easy}} = \frac{8}{g \mu_B} (g \mu_B H Z^2 + 2K_2 \langle S^z \rangle + 4J \langle S^z \rangle)$. It turns out that $\omega^{\text{easy}}$ monotonously increases as $H$ is increased (not shown in Fig. 2c).

For field along the hard axis, $H = H^X \hat{X}$, the time varying magnetization is $\mathbf{M} = (\hat{x}m_x + \hat{y}m_y) e^{-i\omega t} + \hat{z}g \mu_B \langle S^z \rangle$ (where $x$, $y$ and $z$ denote local axes), while the effective field is

$$
\mathbf{H}_{\text{eff}} = \hat{X}H^X - \hat{Y}4\pi m_y + \hat{Z} \frac{2K_2}{g\mu_B} \left( \langle S^z \rangle \cos \phi - \frac{m_x}{g\mu_B} \sin \phi \right)
$$

(6)

(Expressions the effective field in local coordinates and solving the linearized Bloch torque equations, the resonance frequency is found to display a non monotonic behavior (see Fig. 2c), different for $H^X$ below and above the reorientation field $H^X_c = \frac{2K_2 \langle S^z \rangle}{g\mu_B}$)

$$
\omega^\text{hard}_0 = \gamma \left[ 1 + \frac{4\pi g \mu_B^2}{K_2} \right] \sqrt{\left( \frac{2K_2 \langle S^z \rangle}{g\mu_B} \right)^2 - (H^X)^2}, \quad \text{for } H^X < H^X_c
$$

$$
\omega^\text{hard}_0 = \gamma \left[ H^X + 4\pi g \mu_B \langle S^z \rangle \right] \sqrt{(H^X - \frac{2K_2 \langle S^z \rangle}{g\mu_B})^2}, \quad \text{for } H^X > H^X_c
$$

(7)
where \( \langle S^z \rangle = SL(x) \), \( x = x^{hard} = \frac{S}{k_B T} (g \mu_B H^X \sin \phi + 2K_2 \langle S^z \rangle \cos^2 \phi + 4J \langle S^z \rangle) \) and \( \phi \) is given by the equilibrium condition, Eq. \( 2 \). It is worth noticing that the finite temperature mean field theory predicts a vanishing resonance frequency of the uniform mode just at \( H_c^X \) as a consequence of the continuous in-plane reorientation of the magnetization.

### C. High temperature expansions

The behavior of spin wave energies in the high temperature regime above \( T_C \) is made particularly interesting by the reduced dimensionality of the system. In two dimensional magnets, the spin system is highly susceptible to perturbations and can respond strongly even at temperatures above \( T_C \). This means that a two dimensional ferromagnet in the paramagnetic regime can still produce large scale correlations when in a static applied field. Furthermore, these correlations are sensitive to the orientation of the applied field relative to the symmetry axis of the anisotropy.

For temperatures above \( T_C \) a finite magnetization is possible only in the presence of an applied field. Furthermore, since the reorientation field is found to vanish at \( T = T_C \), the field-induced magnetization will align parallel to the applied field. A simple estimate of the resonance frequencies above \( T_C \) can thus be made expanding the Langevin function for high temperatures (small \( x \)) \( L(x) \approx \frac{x}{3} - \frac{x^3}{45} \). For field along the easy axis (\( \mathbf{H} = H^X \mathbf{X} \)) and along the hard axis (\( \mathbf{H} = H^X \mathbf{X} \)), respectively, the resulting resonance frequency is

\[
\omega_0^{easy} = \gamma H^Z \left[ 1 + \left( 4\pi g \mu_B + \frac{2K_2}{g \mu_B} \right) \Gamma_+(T) \right]^{1/2} \times \left[ 1 + \frac{2K_2}{g \mu_B} \Gamma_+(T) \right]^{1/2},
\]

\[
\omega_0^{hard} = \gamma H^X \left[ 1 - \frac{2K_2}{g \mu_B} \Gamma_-(T) \right]^{1/2} \left[ 1 + 4\pi g \mu_B \Gamma_-(T) \right]^{1/2}.
\]

with \( \Gamma_\pm(T) = \left( \frac{1}{3} \pm \frac{4K_2}{3g \mu_B k_B T} \right) \frac{g \mu_B S^2}{k_B T} \). The frequencies have a linear dependence on field with different temperature dependent slopes. The slope is smaller for the field in the hard direction because in that case the anisotropy reduces the torque acting on the spins, thereby reducing the susceptibility with respect to the easy case.

### III. GREEN’S FUNCTION THEORY

Although mean field theory has proved useful to study reorientation transitions in multilayer films, it is well known that it grossly underestimates the effect of single site anisotropy in Heisenberg models and that it leads to estimates of the critical temperature generally too high due to the neglect of spin correlations. As the reorientation field depends on anisotropy and on the temperature dependence of magnetization, one can expect that mean field theory is not capable of providing results that can be compared quantitatively with experiments in high quality epitaxial systems. A Green’s function theory using the random phase approximation appears to be a better choice since it provides a more correct treatment of both issues, namely single site anisotropy and spin correlations.

Recently, a number of Green’s function theories were proposed by different authors to treat the problem of the field-induced transition in ultrathin ferromagnetic films. A thorough discussion of the limits of the approximations used in Ref. \cite{14} to treat the reorientation transition was provided by the authors of Ref. \cite{18} who proposed a theory based on a generalization of the Callen decoupling valid for an arbitrary direction of the external field.

In this work we present a Green’s function approach which was developed independently from Ref. \cite{18}. We start from the outset for rotation of the quantization axis for fields below the reorientation transition: i.e., we write the Green’s functions equations of motion in the local reference frame. Next, by a careful treatment of the uniaxial anisotropy through a generalized Callen decoupling \cite{18} of the higher order Green’s functions, we obtain - in a very simple way even in the case of field applied along the hard axis - both the equilibrium condition and the frequency of the magnetic excitations at finite temperature \( T > 0 \). As a consequence of the generalized Callen’s decoupling, the validity of our theory is limited to low values of the ratio \( r = K_2/J \) between the uniaxial anisotropy and the exchange constant. For high values of \( r \), it is advisable to use a more refined Green’s function approach, where the single ion anisotropy terms are treated exactly by introducing higher order Green’s functions and, taking advantage of relations between products of spin operators, a closed set of \( 2S \) equations of motion is obtained for the anisotropy Green’s functions. Also the quantum Monte Carlo approach in Ref. \cite{20} is expected to be appropriate for large values.
of the ratio \( r \), while, in the opposite limit of small \( r \), finite size effects strongly influence the ‘critical region’ around the reorientation transition).

Coming back to our Green’s function approach, we observe that complete agreement with Ref. \[18\] is obtained for the equilibrium condition \[22\] while different expressions are found for the energies of the magnetic excitations \[22\]. Apparently, this discrepancy is due to the fact that in the Green’s function theory of Ref. \[18\] some extra approximation was made, consisting in the treatment of the anisotropy in terms of an effective field, as far as the magnetization and the magnon energies are concerned.

In order to study the reorientation transition and the field dependence of the magnon energies, we have taken particular care in correctly recovering the results of non interacting spin wave theory (see Appendix B) in the low temperature limit, when the local thermal averages tend to constant values, \(<S^z>\rightarrow S\) and \(<(S^z)^2>\rightarrow S^2\). This limit is not recovered \[23\] by the approach in Ref. \[18\]. In particular, we will show (see later Eq. \[19\]) that the well-known feature \[24\] of a zero frequency of the uniform \((k = 0)\) magnon mode at the reorientation field, predicted also by free spin wave theory (see Appendix B), is correctly recovered by our Green’s function approach in the \( T \rightarrow 0 \) limit.

A. Equilibrium condition and energy of the magnetic excitations

In order to calculate the equilibrium condition and the energies of magnetic excitations in the monolayer described by Eq. \[11\] we use a Green’s function formalism \[23\], supplemented by a random phase approximation (RPA) with a careful treatment of the uniaxial anisotropy through a generalized Callen decoupling \[19\]. Details of the calculation in the absence of magnetic dipole-dipole interactions are given in Appendix A. Hereafter we only quote the final results, where dipole-dipole interactions were included in an approximate way (see Appendix C for details). Taking a rotation procedure of the quantization axis \( z \) (see Fig. 1), we obtain the RPA equilibrium condition in the general case of in-plane field with crystallographic components \( H^X \) and \( H^Z \) (see Eq. \[A7\] in Appendix A)

\[
2K_2 f_S \langle S^z_i \rangle \sin \phi \cos \phi + g \mu_B (H^Z \sin \phi - H^X \cos \phi) = 0
\]

where the factor \( f_S \)

\[
f_S = 1 - \frac{1}{2S^2} [S(S+1) - \langle S_i^z S_i^z \rangle],
\]

is required in order to preserve the kinematic consistency of the magnetic excitations (see later).

It is worth noticing that in the \( T \rightarrow 0 \) limit one has \( \langle S_i^z \rangle \rightarrow S \), so that the RPA equilibrium condition reduces to the equilibrium condition one would obtain in the framework of free spin wave theory, using the standard Holstein-Primakoff transformation for spin operators, by imposing the vanishing of the terms linear in \( a \) and \( a^+ \) in the boson Hamiltonian. The physical meaning of this equilibrium condition is the neglect of correlations between the longitudinal \((z)\) and transverse spin components. In fact, we remind that in the framework of free spin wave theory the longitudinal \((z)\) spin component has a boson representation in terms of zero- and two-boson operators while the transverse \((x, y\) or, equivalently, \(\pm\)) spin components are expressed in terms of one-boson operators (see Appendix B for details). Similarly, the RPA equilibrium condition was obtained by imposing the vanishing of the Green’s function \( G^{ij}_2 (\omega) \) (see Eq. \[A6\] in Appendix A), which correlates the longitudinal and the transverse spin components.

For field applied along the hard axis \((H^Z = 0 \) and \( H^X \neq 0)\), the equilibrium angle is given by \( \phi = \sin^{-1}\left(\frac{g \mu_B H^X}{2K_2 f_S \langle S^z_i \rangle}\right)\) for \( H^X \leq H^X_c \) and \( \phi = \frac{\pi}{2} \) for \( H^X > H^X_c \). The reorientation of the magnetization within the film plane occurs at a critical value of the applied field \( H^X_c = \frac{2K_2 f_S \langle S^z_i \rangle}{g \mu_B} \). Notice that, as in the mean field theory, the reorientation field is a function of temperature, but the RPA condition contains the additional temperature dependent correlation function \( \langle S^z_i S^z_i \rangle \) in the kinematic consistency factor \( f_S \). The latter factor is obtained by a careful decoupling of the higher order Green’s functions coming from the uniaxial anisotropy \[23\] and is required in order to preserve the kinematic consistency of the magnetoexcitations: i.e., for \( S = 1/2 \), the uniaxial anisotropy does not contribute to the magnon energy (see later, Eqs. \[12\] and \[13\]). In fact, for \( S = 1/2 \) one has \( f_S = 0 \); for \( S = 1 \), which is the simplest non-trivial case, one has \( f_S = 1/2 \langle S^z_i S^z_i \rangle \).

The general form of the magnon energies in the momentum representation is, including also the magnetic dipole-dipole interaction (see Appendices A and C for details)

\[
E_{k||} = \sqrt{A_{k||}^2 - B_{k||}^2}
\]

where

\[
A_{k||} = 4J\langle S^z_i \rangle(1 - \gamma_{k||}) + 2K_2 f_S \langle S^z_i \rangle \cos^2 \phi - K_2 f_S \langle S^z_i \rangle \sin^2 \phi + \frac{1}{2} \frac{4\pi w f_w \langle S^z_i \rangle}{g \mu_B (H^X \sin \phi + H^Z \cos \phi)}
\]

\[
B_{k||} = 4J\langle S^z_i \rangle(1 - \gamma_{k||}) + 2K_2 f_S \langle S^z_i \rangle \cos^2 \phi - K_2 f_S \langle S^z_i \rangle \sin^2 \phi + \frac{1}{2} \frac{4\pi w f_w \langle S^z_i \rangle}{g \mu_B (H^X \sin \phi + H^Z \cos \phi)}
\]
FIG. 3: Green’s function calculation of the energy gap for field applied along the hard axis (full lines) and the easy axis (dashed lines) at four different temperatures (T/T_C=0.5, 0.9, 1.05 and 1.2, respectively). With the Hamiltonian parameters used for the calculations, J = 500 K, K_2 = 5 K, 4\pi w f_w = 1 K, S = 1 (giving T_C = 502.4 K), a magnetic anisotropy of the spin wave energies is found to persist up to T/T_C = 1.2.

In Eqs. (13,14), \( \gamma_{k_i} = \frac{1}{2} \left[ \cos(k_x a) + \cos(k_z a) \right] \) is the geometric factor for the simple quadratic (s.q.) lattice and \( w = \frac{(g \mu_B)^2}{a^3} \) denotes the strength of the magnetic dipole-dipole interaction, while the factor \( f_w = 1.078 \) comes from the calculation of dipolar sums for the s.q. lattice. Notice that the wavevector dependence of the dipolar sums was neglected for the sake of simplicity (see the Appendix C for details). Such an approximation for the dipolar interaction is certainly plausible in the presence of a sufficiently large gap in the spectrum.

For S = 1 the relevant thermal averages in the RPA theory are (general expressions for any value of S can be found in Appendix A)

\[
\langle S^z_i \rangle = \frac{1 + 2\Phi(T)}{1 + 3\Phi(T) + 3\Phi^2(T)}
\]

\[
\langle (S^z_i)^2 \rangle = S(S+1) - (1 + 2\Phi(T))\langle S^z_i \rangle = \frac{1 + 2\Phi(T) + 2\Phi^2(T)}{1 + 3\Phi(T) + 3\Phi^2(T)}
\]

where the temperature dependence is self-consistently given by (N is the total number of spins in the s.q. lattice)

\[
\Phi(T) = \frac{1}{2N} \sum_{k_i} \left[ \frac{A_{k_i}}{E_{k_i}} \coth \left( \frac{1}{2} \beta E_{k_i} \right) - 1 \right].
\]
For zero field, in the \( T \to T_C \) limit, one has \( \langle S^z \rangle \to 0 \), so that \( E_{k_0} \) vanishes with it and \( \Phi(T) \) diverges. Thus from Eqs. 15 and 16 one has \( \langle S^z \rangle \to \frac{2}{3\pi f(T)} \) and \( \langle S^z S^z \rangle \to \frac{2}{3} \), respectively. By equating the expression 14 (expanded for \( E_{k_1} \to 0 \)) with \( \Phi(T) = \frac{2}{3\langle S^z \rangle} \), one obtains the following expression for the Curie temperature of the \( S = 1 \) ferromagnetic monolayer in zero field

\[
k_B T_C = \frac{2}{3} \left\{ \frac{1}{N} \sum_{k_1} \frac{4J(1 - \gamma_k) + \frac{4}{3} K_2 + \frac{1}{2} \pi w f_w}{4J(1 - \gamma_k) + \frac{2}{3} K_2 + 4 \pi w f_w} \right\}^{-1}
\]

(18)

B. Discussion

In Fig. 2b we show the self-consistently calculated equilibrium angle \( \phi \), magnetization \( M = \langle S^z \rangle \), and magnetization components \( \langle S^z \rangle = \langle S^z \rangle \cos \phi \) and \( \langle S^X \rangle = \langle S^z \rangle \sin \phi \), as a function of the magnetic field \( H \), applied along the hard axis, for fixed finite temperature \( (T/T_C = 0.5) \). Correspondingly, in Fig. 2d the self-consistently calculated energy gap \( E_0 \) is shown as a function of \( H \).

In contrast with the mean field result shown in Fig. 2a, in the framework of Green’s function theory we find that the orientation angle \( \phi \) varies discontinuously at the reorientation field and that the magnetization modulus \( M = \langle S^z \rangle \) decreases as \( H \) is increased from 0 to the critical value \( H^X_c = \frac{2 K g M (\langle S^z \rangle)}{\gamma \pi a} \). This is related to the more accurate treatment of spin correlations in the Green’s function theory with respect to mean field theory. In fact we observe that both the temperature and field dependence of \( \langle S^z \rangle \) occurs through the quantity \( \Phi(T) \) in Eq. 14, which is determined by a summation over all magnon modes. At fixed temperature, the strongest contribution to \( \Phi(T) \) comes from the \( k = 0 \) mode since this has the lowest energy for \( H^X \to (H^X_c)^{-} \). This explains why \( M = \langle S^z \rangle \) decreases in this limit. In turn, the orientation angle \( \phi \), whose value is determined by the value of \( \langle S^z \rangle \), undergoes an abrupt variation at the reorientation field.

As a consequence of the discontinuity in the orientation angle \( \phi \), the energy gap \( E_0 \) displays a discontinuous jump at the reorientation field \( H^X_c = \frac{2 K g M (\langle S^z \rangle)}{\gamma \pi a} \) (see Fig. 2d) and the energy does not go to zero at the reorientation field. This is in contrast with the finite temperature mean field result displayed in Fig. 2b.

It is important to note that the softening of the \( k = 0 \) magnon energy at the reorientation field \( H^X_c \) is correctly recovered by our Green’s function approach in the \( T = 0 \) limit. In fact for \( T \to 0 \) and \( S = 1 \) one has \( \langle S^z \rangle = \langle S^z S^z \rangle \to 1 \) and the energy of the \( k = 0 \) magnon mode exactly coincides with the result of non interacting spin wave theory for field \( H^X \) applied along the hard direction (see Appendix B for details)

\[
E_0 = \left[ (K)^2 - (g \mu_B H^X)^2 \right]^{1/2} \times \left[ 1 + \frac{4 \pi w f_w}{K} \right]^{1/2}, \text{ for } H^X \leq H^X_c
\]

\[
E_0 = \left[ g \mu_B H^X - K \right]^{1/2} \times \left[ g \mu_B H^X + 4 \pi w f_w \right]^{1/2}, \text{ for } H^X > H^X_c.
\]

(19)

This energy is immediately seen to vanish continuously at the reorientation field \( H^X_c = \frac{K}{g \mu_B} \).

The magnitude of the gap energy at zero field is interesting to consider. The zero field gap value is a measure of the effective anisotropy energy, and is strongly temperature dependent: the zero field gap value vanishes for \( T > T_C \), as well as the reorientation field. This is clearly a consequence of the Callen decoupling10 of the higher order Green’s functions generated by the uniaxial anisotropy, yielding a decoupling factor proportional to the magnetization, see Eq. 14. Physically this is related to the absence of a collectively ordered magnetic state for \( T > T_C \). However, as discussed by Jensen et al.,20 the vanishing of the effective anisotropy for \( T > T_C \) does not indicate that the microscopic anisotropy vanishes either: a single magnetic moment in the paramagnetic state is still subject to the anisotropy even if the net magnetization is zero. While in Ref. 2 such a “paramagnetic anisotropy” was revealed by magnetic susceptibility measurements, in the present paper we rather focus on the gap energy behavior. This feature is illustrated in Fig. 3, where the gap energy is plotted as a function of the magnetic field applied in the easy and in the hard direction, at various fixed temperatures, both below and above \( T_C \). For \( T \geq T_C \), the easy- and hard-direction energies are each zero at zero field, and their field dependence becomes more and more linear as \( T \) increases. However, the calculated slopes are different, in agreement with the mean field argument: the hard axis alignment slope is smaller than the easy axis alignment slope. Finally, it is worth noticing that for a realistic value of the ratio \( K_2/J \) in an ultrathin film (\( \approx \frac{1}{100} \), as in our numerical calculations), the magnetic anisotropy of the energy gap is expected to persist well above \( T_C \) (up to \( T/T_C \approx 1.2 \), see Fig. 3d).
IV. CONCLUSIONS

Using a Green’s function theory with random field approximation we have shown that the field driven reorientation transition of a ferromagnetic monolayer with uniaxial anisotropy is expected to be continuous at $T = 0$ (in agreement with free spin wave theory) and discontinuous at finite temperature. In contrast, mean field theory predicts a field driven reorientation transition continuous at any temperature. Below the critical temperature $T_C$, the Green’s function theory predicts a magnetic excitation gap (the resonance mode) which does not completely soften at the reorientation field.

The question of the order of the transition should be considered as a still open question, because it is not fully clear if and how the decoupling scheme affects that behavior. One step in this direction might be accomplished performing a decoupling of the Green’s functions on a higher level. It would also be interesting to analyze the order of this type of transition in experimental systems (e.g., by ferromagnetic resonance measurements of the uniform mode as a function of the field).

The magnetization of a two dimensional ferromagnet is highly susceptible above the critical temperature $T_C$. This means that a large magnetization can be induced with relatively small applied fields in the paramagnetic regime. Such effects were demonstrated in beautiful experiments on ultrathin Fe/W(110) films by Back et al. More recently, such “paramagnetic anisotropy” effects were investigated by means of magnetic susceptibility measurements on ultrathin Co/Cu(111) films by Jensen et al. In our case, this means that a two dimensional film with well defined anisotropies can support magnetic excitations in the paramagnetic regime with energies that depend upon the orientation of the applied field relative to the anisotropy symmetry axis. We have demonstrated this with our Green’s function theory in the RPA and shown that there can be a sizeable difference in magnetic excitation energies at finite fields. We show that the field dependence of the energy is strongly temperature dependent and different for field alignments along easy and hard directions.

It would be interesting to put our prediction of an anisotropy of the magnetic excitation energies in the paramagnetic phase to an experimental test. This may be possible in ultrathin magnetic films using inelastic light scattering or ferromagnetic resonance measurements.

In fact, recent Brillouin light scattering experiments in Fe/GaAs(100) ultrathin films ($t_{Fe} = 4.5$ Å) showed anisotropy in observed frequencies above some ordering temperature. However, it is not clear from the results whether the film is continuous or is instead a collection of superparamagnetic islands.

Our mean field arguments, for example, apply equally well to paramagnetic films above the Curie temperature and to collections of superparamagnetic particles above the blocking temperature. The only difference might be that a superparamagnetic "film" would be comprised of a distribution of particle sizes, each with a different local internal magnetic field. Resonance in the superparamagnetic system would contain many different frequency contributions and should therefore have a correspondingly broad linewidth. Resonance in the paramagnetic continuous film would have a linewidth determined by higher moments in the spin correlation functions, and should therefore also have a distinctive temperature dependence different from the superparamagnetic case.

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APPENDIX A: GREEN’S FUNCTION FORMALISM

In order to calculate the equilibrium configuration of the monolayer magnetization and the energies of the magnetic excitations, we consider the following Green’s functions

$$G_{ij}^\alpha = \langle \langle S_i^\alpha ; S_j^- \rangle \rangle; \quad \alpha = +, -, z$$

where $S_i^\pm = S_i^x \pm i S_i^y$ and $x, y, z$ denote a reference frame where $z$ is the equilibrium direction of the in-plane magnetization, forming an angle $\phi$ with the easy in-plane direction $Z$ (see Fig. 1). The equations of motion for the time Fourier-transformed Green’s functions $G_{ij}^\alpha (\omega)$ are

$$\omega G_{ij}^\alpha (\omega) = \langle [S_i^\alpha , S_j^-] \rangle + \langle [S_i^\alpha , \mathcal{H}] ; S_j^- \rangle_\omega$$

(A1)

(A2)
where \( \langle A \rangle = \frac{1}{Z} \text{Tr}(e^{-\beta H} A) \) denotes a thermal average (\( \beta = \frac{1}{k_B T} \) and \( Z = \text{Tr}(e^{-\beta H}) \) is the partition function). Taking into account that \( S^X = S^x \cos \phi + S^z \sin \phi \) and \( S^Z = -S^x \sin \phi + S^z \cos \phi \), one obtains

\[
\omega \ G_{ij}^X(\omega) = \langle [S_i^x, S_j^z] \rangle \\
+ \frac{1}{2} J \sum_k \langle [S_k^z - S_k^z, S_i^x - S_i^x] \rangle \omega \\
- \frac{1}{2} K_2 \sin^2 \phi \langle [S_i^x, S_j^z - S_i^x, S_j^z] \rangle \omega \\
+ \frac{1}{2} K_2 \sin \phi \cos \phi \langle [2S_k^x, S_i^x + S_i^x, S_j^z - S_i^s, S_j^z] \rangle \omega \\
+ \frac{1}{2} \mu_B H^Z \sin \phi \langle G^X_{ij}(\omega) - G^Z_{ij}(\omega) \rangle \\
- \frac{1}{2} \mu_B H^X \cos \phi \langle G^X_{ij}(\omega) - G^Z_{ij}(\omega) \rangle
\]  
(A3)

The higher order Green’s functions on the right hand sides of the previous equations have now to be decoupled to obtain a closed set of equations. We start using the naïve RPA decoupling \( \langle H \rangle \) and \( \langle g_{µ} \rangle \), both for \( i \neq k \) and \( i = k \); kinematic consistency corrections will be taken into account later on

\[
\langle [S_i^x, S_k^x] \rangle \omega = \langle S_i^x \rangle G^X_{ij}(\omega) + \langle S_k^x \rangle G^X_{ij}(\omega) + \langle S_i^x \rangle G^X_{ij}(\omega)
\]  
(A5)

Next, taking into account that the local axis \( z \) is, by its own definition, the equilibrium direction of the magnetization, we can safely put \( \langle S^x \rangle \approx 0 \) and \( \langle S^z \rangle \approx 0 \) in the equations of motion of the three Green’s functions \( G^X_{ij}(\omega), G^Z_{ij}(\omega) \) and \( G^Z_{ij}(\omega) \).

The equation of motion for \( G^Z_{ij}(\omega) \) provides the equilibrium configuration. In fact in the previous approximations we obtain

\[
\omega G^Z_{ij}(\omega) = \frac{1}{2} \left[ G^X_{ij}(\omega) - G^Z_{ij}(\omega) \right] \times \left[ K_2 \langle S_i^z \rangle \sin 2\phi + \mu_B (H^Z \sin \phi - H^X \cos \phi) \right]
\]  
(A6)

so that the equilibrium angle \( \phi \) is obtained imposing the vanishing of the term in braces on the r.h.s.

\[
2K_2 \langle S_i^z \rangle \sin \phi \cos \phi + \mu_B (H^Z \sin \phi - H^X \cos \phi) = 0
\]  
(A7)

For field applied along the easy axis (\( H^Z \neq 0 \) and \( H^X = 0 \)), one obviously obtains \( \phi = 0 \); i.e., the magnetization is always directed along the field (\( Z = z \)) direction. For field applied along the hard axis (\( H^Z = 0 \) and \( H^X \neq 0 \)), the equilibrium angle is given by

\[
\phi = \sin^{-1} \left( \frac{\mu_B H^X}{2K_2 \langle S_i^z \rangle} \right), \quad \text{for } H^X \leq H^X_c
\]
\[
\phi = \frac{\pi}{2}, \quad \text{for } H^X > H^X_c
\]  
(A8)

so that a reorientation of the magnetization within the film plane occurs at a critical value of the applied field \( H^X_c = \frac{2K_2 \langle S_i^z \rangle}{\mu_B} \) which turns out to be temperature dependent. For \( H^X > H^X_c \), the magnetization becomes parallel to the external field \( H^X \) applied along the hard direction.
where $F^\pm(k_\parallel) = \frac{1}{N} \sum_{ij} e^{-i\mathbf{k}_\parallel \cdot \mathbf{r}_{ij}} \langle [\pm \mathbf{S}^i_z, \mathbf{S}^j_z] \rangle$ and $\mathbf{k}_{\parallel} = \frac{1}{z_{nn}} \sum_\delta e^{-i\mathbf{k}_\parallel \cdot \delta}$ ($\delta$ are the vectors joining a given lattice site to its $z_{nn}$ nearest neighbors). For a square lattice with lattice constant $a$ one has $z_{nn} = 4$ and the geometrical factor takes the form $\gamma_{\parallel} = \frac{1}{2}[\cos(k_z a) + \cos(k_z a)]$.

The previous equations can be rewritten in a more compact form

$$
\begin{bmatrix}
\omega - A_{k_\parallel} & B_{k_\parallel} \\
-B_{k_\parallel} & \omega + A_{k_\parallel}
\end{bmatrix}
\begin{bmatrix}
G^+(\omega, k_\parallel) \\
G^-(\omega, k_\parallel)
\end{bmatrix}
= \begin{bmatrix}
F^+(k_\parallel) \\
F^-(k_\parallel)
\end{bmatrix}
$$

(A10)

where the quantities $A_{k_\parallel}$ and $B_{k_\parallel}$ take the following expressions

$$
A_{k_\parallel} = 4J(S_i^z)(1 - \gamma_{k_\parallel}) + 2K_2(S_i^z) \cos^2 \phi - K_1(S_i^z) \sin^2 \phi + \mu_B(H^Z \cos \phi + H^X \sin \phi)
$$

$$
B_{k_\parallel} = K_2(S_i^z) \sin^2 \phi
$$

(A11)

The energies of the magnetic excitations with respect to the equilibrium state are then given by

$$
E_{k_\parallel}^2 = (\hbar \omega_{k_\parallel})^2 = A_{k_\parallel}^2 - B_{k_\parallel}^2
$$

(A12)

It is worthwhile noticing that in the zero temperature limit one has $\langle S_i^z \rangle \rightarrow S$ so that the free spin wave frequency is correctly recovered. In particular, in the $T = 0$ limit we are able to recover the well-known feature of a zero frequency mode for field applied along the hard axis and equal to the critical reorientation field, in contrast with the results obtained by the Green’s function approach in Ref. 15. It is worth noticing that other Green’s function approaches were not able to recover such a feature, either.

So far, in our calculations, the problem of the kinematic consistency of the magnetic excitations was neglected since the naive RPA decoupling was performed also for the Green’s functions coming from the anisotropy term in the Hamiltonian. In the case of field applied along the easy axis ($H^Z \neq 0, H^X = 0$), it is well known that a more correct treatment of the anisotropy term is obtained by using the Anderson-Callen decoupling to decouple the equal site $(i = k)$ Green’s functions

$$
\langle [S_i^\pm S_i^z + S_i^z S_i^\pm; S_j^z] \rangle \omega \approx 2 \langle S_i^z \rangle G_{ij}^\pm(\omega) \times \left\{ 1 - \frac{1}{2S^2} [S(S + 1) - \langle S_i^z S_i^z \rangle] \right\}
$$

(A13)

In this way, for $S = 1/2$, the uniaxial anisotropy does not contribute to the frequency of the excitations, as expected, since for $S = 1/2$ the anisotropy term in the Hamiltonian is a constant. In the case of field applied along the hard axis ($H^Z = 0, H^X \neq 0$) or in the general case ($H^Z \neq 0$ and $H^X \neq 0$), we assume the same prescription for kinematic consistency to be valid as in the easy case, i.e. we replace

$$
K_2(S_i^z) \rightarrow K_2(S_i^z) \left\{ 1 - \frac{1}{2S^2} [S(S + 1) - \langle S_i^z S_i^z \rangle] \right\}
$$

(A14)

both in the equation providing the equilibrium condition and in the equations giving the energies of the magnetic excitations. In the case $S = 1$ considered in this paper, the prescription for kinematic consistency reads simply

$$
2K_2(S_i^z) \rightarrow K_2(S_i^z)/\langle S_i^z S_i^z \rangle
$$

(A15)

In order to determine the average $\langle S_i^z S_i^z \rangle$, we follow the method exposed by Callen in his paper. In addition to the Green’s functions $G_{ij}^\pm = \langle [S_i^\pm S_j^z] \rangle$, we consider the Green’s functions $G_{ij,a}^\pm = \langle [e^{a S_i^z S_i^\pm} S_j^z] \rangle$ where $a$ is a parameter. The $G$’s are found to satisfy the equations of motion

$$
\begin{bmatrix}
\omega - A(k_\parallel) & B(k_\parallel) \\
-B(k_\parallel) & \omega + A(k_\parallel)
\end{bmatrix}
\begin{bmatrix}
G_+^a(\omega, k_\parallel) \\
G_-^a(\omega, k_\parallel)
\end{bmatrix}
= \begin{bmatrix}
F_+^a(k_\parallel) \\
F_-^a(k_\parallel)
\end{bmatrix}
$$

(A16)
which differ from the equation of motion for the \( G \)'s only for the terms \( \mathcal{F}^\pm_a \) on the r.h.s., defined as \( \mathcal{F}^\pm_a (k_\parallel) = \frac{1}{2 \pi} \sum_{ij} e^{-ik_\parallel \cdot r_{ij}} \langle [S^i_j, e^{a S^j_i} S^i_j] \rangle \). Thus from the spectral theorem

\[
\langle BA \rangle = \frac{i}{2 \pi} \lim_{\epsilon \to 0} \int_{-\infty}^{+\infty} \frac{d\omega}{e^{\beta \omega} - 1} \left[ \langle \langle A; B \rangle \rangle (\omega + i \epsilon, k_\parallel) - \langle \langle A; B \rangle \rangle (\omega - i \epsilon, k_\parallel) \right]
\]  

(A17)

applied to \( G^+ \) and \( G^+_a \) one can obtain, respectively, the thermal averages

\[
\langle S^i_j S^j_i \rangle = G^+(k_\parallel) \Phi(T)
\]

\[
\langle e^{a S^i_j} S^j_i \rangle = \mathcal{F}^+_a (k_\parallel) \Phi(T)
\]

(A18)

where

\[
\Phi(T) = \frac{1}{2N} \sum_{k_\parallel} \left[ A_{k_\parallel} / E_{k_\parallel} \right] \coth \left( \frac{1}{2} \beta E_{k_\parallel} \right) - 1
\]

(A19)

For the two-spin correlation function we obtain

\[
\langle S^i_j S^j_i \rangle = 2 \langle S^i_j \rangle \Phi(T)
\]

(A20)

Following Callen, it is now possible to find a second-order differential equation for \( \mathcal{F}^+_a (k_\parallel) \) as a function of the parameter \( a \) that, when solved with opportune boundary conditions, allows to obtain the magnetization as a function of \( \Phi(T) \)

\[
\langle S^i_j \rangle = \frac{[S - \Phi(T)][1 + \Phi(T)]^{2S+1} + [S + 1 + \Phi(T)][\Phi(T)]^{2S+1}}{[1 + \Phi(T)]^{2S+1} - [\Phi(T)]^{2S+1}}
\]

(A21)

and finally

\[
\langle S^i_j S^j_i \rangle = S(S + 1) - \langle S^i_j S^j_i \rangle - \langle S^i_j \rangle = S(S + 1) - (1 + 2\Phi(T))\langle S^i_j \rangle
\]

(A22)

**APPENDIX B: FREE SPIN WAVE THEORY**

We think it useful to briefly present here the results of non interacting spin wave theory, both for the equilibrium condition and the energy of the magnetic excitations in an anisotropic monolayer described by the microscopic Hamiltonian \( \mathcal{H}_c \), since in the \( T \to 0 \) limit the spin wave results are expected to be exactly reproduced by Green’s function theory. Denoting by \( x, y, z \) the local reference frame where \( z \) is the equilibrium direction of the in-plane magnetization, forming an angle \( \phi \) with the easy in-plane direction \( Z \) (see Fig. 1), the local spin components are expressed in terms of boson operators via the Holstein-Primakoff transformation

\[
S^j_i = \sqrt{2S} \left[ 1 - \frac{a^+_j a_j}{2S} \right] a_j, \quad S^j_i = \sqrt{2S} a^+_j \left[ 1 - \frac{a^+_j a_j}{2S} \right], \quad S^j_0 = S - a^+_j a_j
\]

(B1)

while the spin components in the crystallographic reference frame are obtained from the local ones through the equations \( S^X = S^x \cos \phi + S^z \sin \phi \) and \( S^Z = -S^x \sin \phi + S^z \cos \phi \). In the following, for the sake of simplicity, we neglect both dipolar interactions and kinematic consistency corrections to the anisotropy term in Eq. \( \mathcal{H}_c \). Substituting in \( \mathcal{H}_c \), expanding the square roots in Eq. \( \mathcal{H}_c \) (such an approximation is valid for low values of the occupation number \( n_j = a^+_j a_j \), that is for low temperatures), and keeping only up to quadratic terms in the boson operators one obtains, after a space Fourier transformation \( a_j = \frac{1}{\sqrt{N}} \sum_{k_\parallel} a_{k_\parallel} e^{-ik_\parallel \cdot r_{ij}} \)

\[
\mathcal{H} \approx \mathcal{H}_1 + \mathcal{H}_2 = \sqrt{S} \sum_2 \left( a_{k_\parallel} + a^+_k \right) \left[ 2K_2 S \sin \phi \cos \phi + g\mu_B \left( H^Z \sin \phi - H^X \cos \phi \right) \right] + \sum_1 \left[ A_{k_\parallel} a_{k_\parallel} a_{k_\parallel} + \frac{1}{2} B_{k_\parallel} \left( a_{k_\parallel} a_{-k_\parallel} + a^+_k a^+_k \right) \right]
\]

(B2)
where

\[
A_{k||} = 4JS(1 - \gamma_k) + 2K_2S \cos^2 \phi - K_2S \sin^2 \phi + g\mu_B (H^X \sin \phi + H^Z \cos \phi)
\]

\[
B_{k||} = -K_2S \sin^2 \phi.
\]

The equilibrium condition is obtained by imposing the vanishing of the one-boson Hamiltonian \(H_1\)

\[
2K_2S \sin \phi \cos \phi + g\mu_B (H^Z \sin \phi - H^X \cos \phi) = 0
\]

It is readily seen that the spin wave theory result Eq. (19) is reproduced by Green’s function theory in the limit \(T \to 0\), see Eq. (A4).

The two-boson Hamiltonian \(H_2\) can be readily diagonalized and the energy of the non-interacting spin wave excitations turns out to be

\[
E_{k||} = \sqrt{A_{k||}^2 - B_{k||}^2} \tag{B5}
\]

Taking into account both dipolar interactions and kinematic consistency corrections for field \(H^X\) applied along the hard in-plane direction one finally obtains the free spin wave energy explicitly reported in Eq. (19).

APPENDIX C: CONTRIBUTION OF DIPOlar INTERACTIONS

The inclusion of magnetic dipole-dipole interactions (i.e., the last term in Eq. (11) in the equations of motion for the Green’s functions leads to a considerable amount of additional calculations. As they are tedious but straightforward, hereafter we only quote the final result after RPA decoupling. First we consider the commutator of \(H_{dip}\) with \(S^z_i\) in order to investigate the contribution of the dipolar interaction to the equilibrium condition. We have (\(w = \frac{g\mu^2}{a^2}\))

\[
\langle\langle [S^z_i, H_{dip}]; S^z_j \rangle\rangle \omega \approx \frac{3}{2} w \sum_{k \neq l} \{ (S^z_k) \delta_{ik} + (S^z_l) \delta_{il} \}
\]

\[
\{ \left[ (d^{XX}_{kl} - d^{ZZ}_{kl}) \sin \phi \cos \phi + d^{XZ}_{kl} (\cos^2 \phi - \sin^2 \phi) \right] \times \frac{G_{ij}^+(\omega) - G_{ij}^-(\omega)}{2i} \}
\]

where the dipolar sums are defined as (\(\alpha, \beta = X, Y, Z\))

\[
D^{\alpha\beta}(k||) = \sum_{i \neq j} d^{\alpha\beta}_{ij} e^{ik||} \cdot r_{ij} = \sum_{i \neq j} \frac{a^3}{r_{ij}^3} \frac{r_{ij}^\alpha r_{ij}^\beta}{r_{ij}^2} e^{ik||} \cdot r_{ij} \tag{C2}
\]

For a monolayer with the crystallographic \(Y\) axis normal to the film plane one has \(r_{ij}^Y = 0\) so that \(D^{XY} = D^{YZ} = 0\); these relations are approximately satisfied even in an ultrathin film. Moreover, in a lattice with a center of inversion symmetry one has \(D^{XX} = D^{ZZ}\) and \(D^{XZ} = 0\). Thus, as expected, it is proved that the dipolar interaction does not contribute to the equilibrium condition of an in-plane magnetized ultrathin film.

Next, we consider the commutator of \(H_{dip}\) with \(S^z_i\) in order to obtain the contribution of the dipolar interaction to the magnon energies. In the same approximations as before we obtain, after a Fourier transformation

\[
A^{dip}_{k||} = \frac{1}{2} w \langle S^z_i \rangle \sum_{i \neq j} \frac{a^3}{r_{ij}^3} e^{ik||} \cdot r_{ij} \times \left\{ 1 + 3 \left( \frac{r_{ij}^X \cos \phi + r_{ij}^Z \sin \phi}{r_{ij}} \right)^2 \right\}
\]

\[-2 \left\{ 1 - 3 \left( \frac{r_{ij}^X \cos \phi + r_{ij}^Z \sin \phi}{r_{ij}} \right)^2 \right\} \]

\[
B^{dip}_{k||} = \frac{1}{2} w \langle S^z_i \rangle \sum_{i \neq j} \frac{a^3}{r_{ij}^3} e^{ik||} \cdot r_{ij} \times \left\{ 1 - 3 \left( \frac{r_{ij}^X \cos \phi - r_{ij}^Z \sin \phi}{r_{ij}} \right)^2 \right\} \tag{C3}
\]

In order to simplify the numerical calculations, we neglect the dependence of \(A^{dip}_{k||}\) and \(B^{dip}_{k||}\) on \(k||\). Taking into account that for a two-dimensional lattice with a center of inversion symmetry one has

\[
\sum_{i \neq j} \frac{a^3}{r_{ij}^3} \left( \frac{r_{ij}^X}{r_{ij}} \right)^2 = \sum_{i \neq j} \frac{a^3}{r_{ij}^3} \left( \frac{r_{ij}^Z}{r_{ij}} \right)^2 = -\frac{1}{2} \sum_{i \neq j} \frac{a^3}{r_{ij}^3} = -\frac{4}{3} \pi f_w \tag{C4}
\]
with $w = 1.078$ for the s.q. lattice, we thus obtain the approximate expressions \cite{13} and \cite{14} reported in Section II. Clearly, such an approximation on the dipolar part of the Hamiltonian is allowed as long as the strength of the dipolar interaction $w$ is smaller than the intensity of the uniaxial anisotropy and/or the Zeeman term.

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1. C. H. Back, A. Kashuba, D. Pescia, Phys. Low-Dim. Struct. 2, 9 (1994).
2. P. J. Jensen, S. Knappmann, W. Wulfhekel, H. P. Oepen, Phys. Rev. B 67, 184417 (2003).
3. S. O. Demokritov, N. M. Kreines, V. I. Kudinov, S. V. Petrov, Sov. Phys. JETP 68, 1277 (1989).
4. J. R. Dutcher, B. Heinrich, J. F. Cochran, D. A. Steigerwald, W. F. Egelhoff, Jr., J. Appl. Phys. 63, 3464 (1988).
5. T. Holstein and H. Primakoff, Phys. Rev. 58, 1098 (1940).
6. Y. Yafet, J. Kwo, E. M. Gyorgy, Phys. Rev. B 33, 6519 (1986).
7. P. Bruno, Phys. Rev. B 43, 6015 (1991).
8. P. Politi, A. Rettori, M. G. Pini, J. Magn. Magn. Mater. 113, 83 (1992).
9. M. G. Pini, A. Rettori, D. Pescia, N. Majlis, S. Selzer, in Microscopic Aspects of Nonlinearity in Condensed Matter, edited by A. R. Bishop, V. L. Pokrovsky and V. Tognetti (Plenum, New York, 1991); M. G. Pini, A. Rettori, D. Pescia, N. Majlis, S. Selzer, Phys. Rev. B 45, 5037 (1992).
10. In this geometry the thermal average in the field direction is $\langle S^X \rangle$. This average does not appear in the effective field in the case of a continuous thin film, but could appear if additional in plane shape factors existed.
11. A. Hucht and K. D. Usadel, Phil. Mag. B 80, 275 (2000). K. D. Usadel and A. Hucht, Phys. Rev. B 66, 024419 (2002).
12. J. F. Devlin, Phys. Rev. B 4, 136 (1971).
13. D. N. Zubarev, Usp. Fiz. Nauk. 71, 71 (1960) [Sov. Phys. Usp. 3, 320 (1960)].
14. P. Fröbrich, P. J. Jensen, P. J. Kuntz, Eur. Phys. J. B 13, 477 (2000).
15. P. Fröbrich, P. J. Jensen, P. J. Kuntz, A. Ecker, Eur. Phys. J. B 18, 579 (2000).
16. P. Fröbrich, P. J. Kuntz, J. Phys.: Condens. Matter 16, 3453 (2004).
17. P. Fröbrich, P. J. Kuntz, S. Saber, Ann Phys. (Leipzig) 11, 387 (2002).
18. S. Schwieger, J. Kienert, W. Nolting, Phys. Rev. B 71, 024428 (2005).
19. H. B. Callen, Phys. Rev. 130, 890 (1963); F. B. Anderson, H. B. Callen, Phys. Rev. 136, A1068 (1964).
20. P. Henelius, P. Fröbrich, P. J. Kuntz, C. Timm, P. J. Jensen, Phys. Rev. B 66, 094407 (2002).
21. Compare Eq. (18) of Ref. 15 with Eq. (19) of the present paper.
22. Compare Eq. (19) of Ref. 15 with Eqs. (12), (13), (14) of the present paper.
23. In fact, from Eq. (19) of Ref. 15 one has that for zero wavevector the magnon energy reduces to the effective field $B$ so that, substituting the condition for the magnetization angle obtained from Eq. (18) of Ref. 15 one finds that, in the $T \to 0$ limit (when the thermal averages $\langle S_x \rangle$ and $\langle S^x \rangle$ tend to constant values), the magnon energy does not vanish at the reorientation field.
24. A. H. Morrish, The Physical Principles of Magnetism, (Wiley, New York, 1965), p. 555.
25. I. Junger, D. Ihle, J. Richter, A. Klümpner, Phys. Rev. B 70, 104419 (2004).
26. G. Carlotti et al., private communication (2004).
27. U. Balucani, V. Tognetti, M. G. Pini, J. Phys. C.: Solid State Phys. 12, 5513 (1979).