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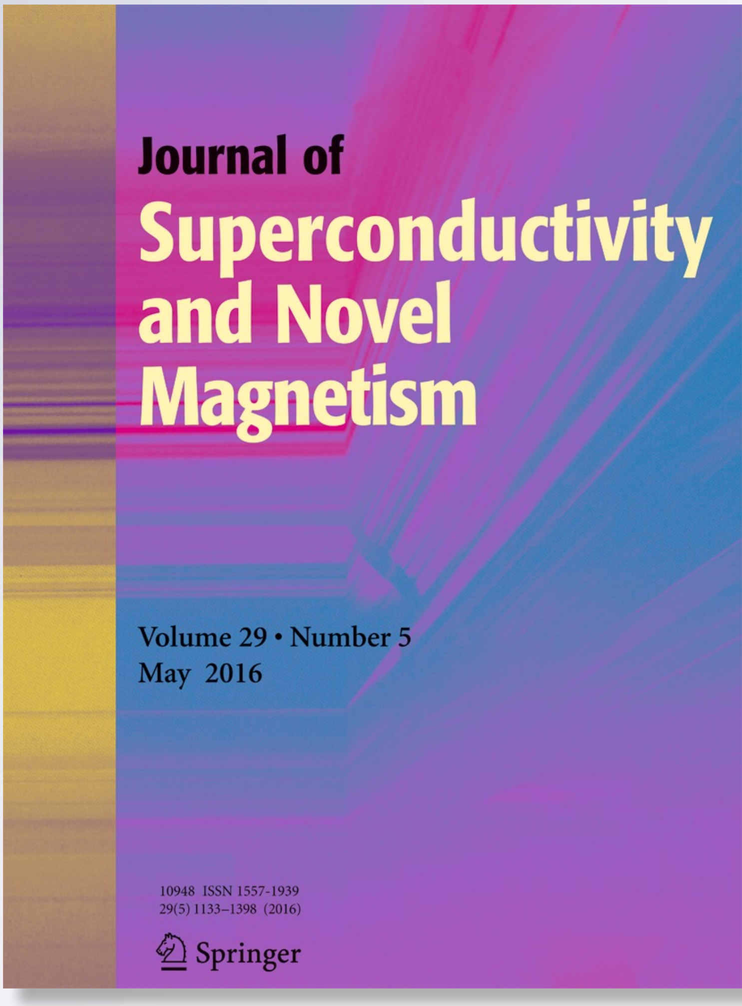

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ORIGINAL PAPER

Quantum Theory of Spin Wave Gap in Ultrathin Magnetic Films

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Abstract A simple quantum model is presented for the spin wave energy gap in single-layer and thin magnetic films that include both the magnetic out-of-plane and in-plane anisotropies. The films are assumed to be under the influence of the out-of-plane direction of the applied magnetic field at zero temperature. The calculated equations present a nonzero spin wave gap at zero magnetic field which is strongly affected by anisotropies. The effects of the film thickness and the role of the applied field are also examined. We discuss the results in connection with experimental data reported for nanocrystalline amorphous CoFeB films with growth-induced anisotropy.

Keywords Magnetic anisotropy · Spin wave gap · Ultrathin film

1 Introduction

Experiments on the phenomenon of magnetic anisotropy which is closely related to the utilization of the direction of magnetization in thin magnetic films and multilayer structures have long been an interesting topic for new technologies [1, 2]. The inclusion of magnetic anisotropy plays a key role and leads to an energy gap in the spin wave spectrum in such materials. A large number of experimental and theoretical works have been performed using ultrathin films with thickness of only a few atomic layers or multilayers. A good example for this phenomenon is in layered magnetic structures [3] and a single monolayer and for thin films consisting of two or three atomic layers [4–10], ferromagnetic nanowires [11], epitaxial Fe-deficient yttrium iron garnet (YIG) films [12], nanocrystalline CoFeB film [13], and so on.

Under the influence of an applied magnetic field, the behavior of magnetic materials exhibits a strong surface anisotropy and may lead to an easy axis for the magnetization normal to the film planes. The shape anisotropy arises from the long-range magnetic dipolar interaction which favors an in-plane orientation of the magnetization, but the magnetocrystalline anisotropy due to the spin orbit interaction shows clearly much more complicated behaviors in thin films, in such a way that with increasing film thickness a reorientation of the magnetization must occur, from perpendicular to in plane as the temperature is increased [14–18].

In our previous works [19, 20], we have calculated the spin wave gap of two-dimensional magnets at zero temperature in an existence of applied magnetic field which was perpendicular/in to the plane. The magnetic anisotropy contribution and the direction of the applied field strongly influence the spin wave gap of an ultrathin film with small thickness.

The purpose of this paper is to present a simple quantum model which gives the spin wave gap as a function of the applied magnetic field which is perpendicular to the film plane at zero temperature. We concentrate on the regime where both the second-order perpendicular anisotropy term of the film and the uniaxial in-plane anisotropy which breaks the fourfold symmetry may provide comparable contributions to the dynamical processes. We use two types

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of the dimensionless quantities which are in-plane to perpendicular anisotropy and magnetic field to perpendicular anisotropy in order to demonstrate how the spin wave energy gap condition varies with the influence of these parameters. The roles of these parameters are also examined by means of the number of atomic layers.

2 Basic Expressions

We first consider the effect upon the spin wave spectrum of an anisotropy within the easy plane. The field dependence of the spin wave gap in a nanocrystalline CoFeB film was studied in a wide frequency range. Critical spin fluctuations in the form of soft modes were observed in a whole range of orientations of the magnetic field perpendicular to the easy magnetic axis [13]. For a film in the (x, y) plane, we assume that the total Hamiltonian based on using a simplified approach neglecting exchange energy can be expressed generally as follows:

$$\mathcal{H} = \mathcal{H}_a + \mathcal{H}_z \tag{1}$$

The first term of (1) represents the magnetic anisotropy energy. We have included the out-of-plane anisotropy, K_o, which includes the shape anisotropy term and an in-plane anisotropy, K_p, which breaks the fourfold symmetry so that the film is a cubic material, with the z axis being taken as the surface normal. All of the anisotropy constants depend

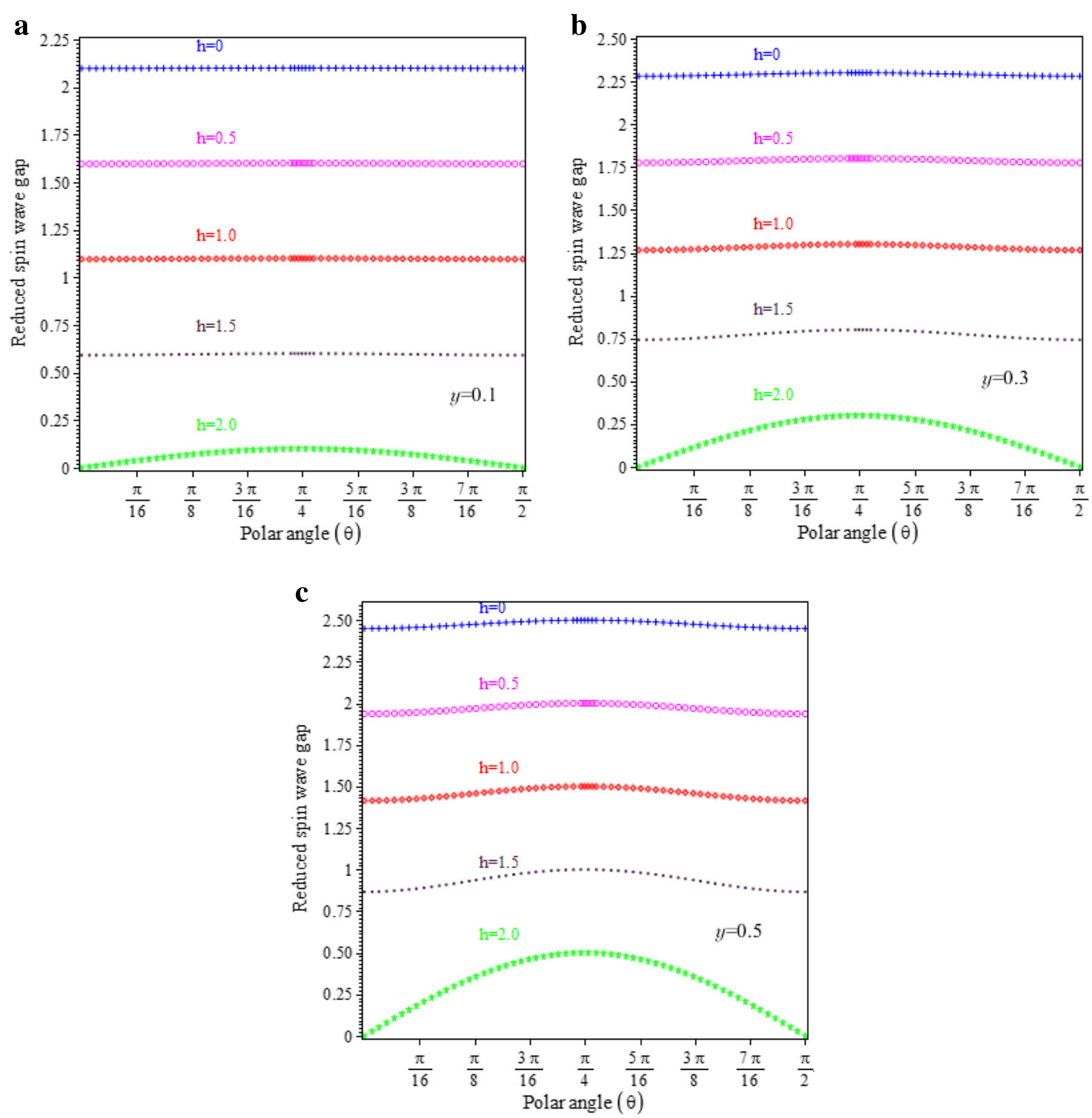

Fig. 1 Reduced spin wave gap as a function of the polar angle θ for different reduced external magnetic fields and values of y: **a** $y = 0.1$, **b** $y = 0.3$, and **c** y=0.5

on thickness L. The second term of (1) implies the Zeeman energy of the spins in a magnetic field of magnitude H which throughout we suppose is directed along the z axis. Equation (1) can be defined with respect to some local axes (λ, μ). We use the transformation between the local axis and the axis defined by the anisotropy [21]:

$$S_\lambda = S_y \sin\theta + S_z \cos\theta$$
$$S_\mu = S_y \cos\theta - S_z \sin\theta.$$

The total spin wave Hamiltonian can be written in the following form:

$$\begin{aligned}\mathcal{H} = \Sigma_n \Big\{ & K_\circ(n) S_z^2(n) - K_p(n) \\ & \times \left[S_\lambda^2(n) - \left(S_\lambda^2(n) - S_\mu^2(n) \right) \sin^2\theta \right. \\ & \left. - S_\lambda(n) S_\mu(n) \sin 2\theta \right] - g\mu_B H S_z(n) \Big\}. \end{aligned} \quad (2)$$

To examine the spin wave gap at zero temperature, we use the Holstein-Primakoff transformation [22]. According to our choice of coordinate axes, the components of a spin vector $S(n)$ are represented in terms of boson creation and annihilation operators $a_n^\dagger$ and a_n by the following expressions:

$$S_x(n) = \frac{\sqrt{2S}}{2}\left(a_n + a_n^\dagger\right), \; S_y(n) = \frac{\sqrt{2S}}{2i}\left(a_n - a_n^\dagger\right),$$
$$S_z(n) = S - a_n^\dagger a_n.$$

The Hamiltonian (2) can be rewritten in the following form:

$$\begin{aligned}\mathcal{H} = \Sigma_n \Big\{ & K_o(n) S_z^2(n) - K_p(n) \left[S_y^2(n) + \left(S_x^2(n) - S_y^2(n) \right) \sin^2\theta \right. \\ & \left. - S_x(n) S_y(n) \sin 2\theta \right] - g\mu_B H S_Z(n) \Big\} \end{aligned} \quad (3)$$

where g is the gyromagnetic ratio and μ_B is the Bohr magneton. The spin operators are S_x, S_y and S_z, and they are defined by their commutation relations $[S_x, S_y] = i\hbar S_z$ and so on [23], and at low temperature, $S_z \approx S$. The Hamiltonian (3) is diagonalized by the introduction of spin operator S_α where $\alpha = x, y$ is chosen to satisfy the relation $[S_\alpha, \mathcal{H}] = \omega_\alpha S_\alpha$ [24]. The eigenfrequencies are the roots of

$$\det \begin{vmatrix} \omega - A & -B \\ B & \omega + A \end{vmatrix} = 0,$$

or

$$\omega = (A^2 - B^2)^{1/2}. \quad (4)$$

We now consider several special cases. If the anisotropy terms are zero, we have

$$\omega_o = g\mu_B (H + 4\pi M_s) \quad (5)$$

where M_s is the saturation magnetization. Equation (5) is in agreement with the classical result except for the exchange energy [25]. For general angles θ between the z axis and vector S_z, the secular equation is given by (4) with

$$A = g\mu_B H - 2K_\circ - 2K_p \cos^2\theta$$

and

$$B = g\mu_B H - 2K_\circ - 2K_p \sin^2\theta.$$

It is useful to introduce the anisotropic fields $K_\circ = g\mu_B H_o$ and $K_p = g\mu_B H_p$. For $\theta = 0$ and $H \geq H_o + H_p$, we find (with $S = 1$) the following expression:

$$\omega_k = g\mu_B [(H - 2H_o)(H - 2H_o - 2H_p)]^{1/2} \quad (6)$$

which is not zero if $H = 0$ as we have expected. Equation (6) is the same as in ref. [13], except for the H_o which includes the shape anisotropy term. Manuilov et al. [13] have measured the in-plane anisotropy field and gyromagnetic g-factor as a function of two different orientations of the external magnetic field: perpendicular to the easy and hard axes. In the computation, $H \leq 10^4$ Oe, $H_p = 535$ Oe, the negative out-of-plane anisotropy $H_o \approx -1$ kOe and $g = 2.09$ for the 4-μm-thick $(CoFeB)_{1-x}-(SiO_2)_x$ with $x = 0.235$. Figure 1a–c shows the spin wave gaps of ultrathin film as a function of the out-of-plane angle θ for the dimensionless parameters $y = \frac{H_p}{H_o}$ and $h = \frac{H}{H_o}$. As can be seen, at $h = 0$, the spin wave gap is essentially determined by the magnetic anisotropy. Obviously, the spin wave gap decreases with increasing magnetic fields. As y increases, the directional dependence of the spin wave gap also inreases periodically and the spin wave gap approaches that of (6). The anisotropy modes are significantly affected by changing the orientation of the magnetization with respect to the crystallographic axes.

Figure 2 shows the plot of the reduced spin wave gap as a function of the reduced applied magnetic field with

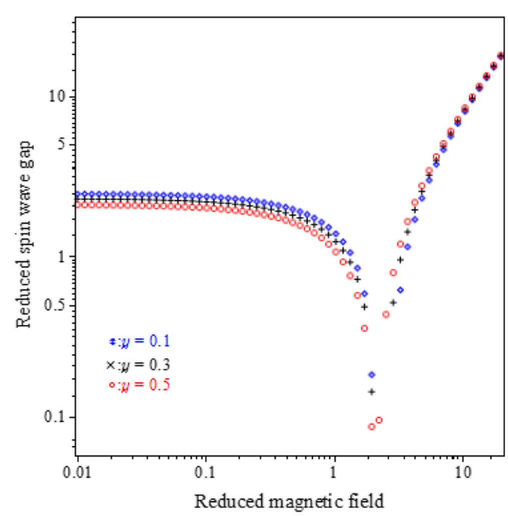

Fig. 2 Reduced spin wave gap as a function of the reduced applied magnetic field with respect to the [001] axis for various values of y

respect to the [001] axis for various values of y. As it is seen, the spin wave gap calculated at zero temperature is almost constant below about $h \leq 0.1$ and displays at a critical field value of $h \simeq 2$. In this case, the contribution of the Zeeman energy to the spin wave gap is overwhelmed by the demagnetizing energy and the curves decrease slowly with increasing field. Note that the modes have a very sharp minimum but they do not vanish. We interpret this as a consequence of nonuniform magnetization. This points out that an easy plane magnet is unstable in two dimensions, and hence, any small effect which produces stabilization has a dramatic influence. This may be due to strong film distortions which contribute to the spin wave gap. At larger values of the critical field, the gap is found to increase almost linearly in which the three lines exactly coincide. This indicates that the film is uniformly magnetized. A qualitatively similar behavior has been observed in ferromagnetic nanowires [11] and ferromagnetic films as well as multilayers with large out-of- plane anisotropy [3, 5]. The in- and out-of-plane anisotropies should dominate in determining the spin wave gap. It is important to rule out the possible effect of an applied magnetic field in determining the spin wave gap which validates the presented models.

We next concentrate to achieve a better understanding of the nature of the involved anisotropies which depend on the film thickness L. Physically, the experimental data

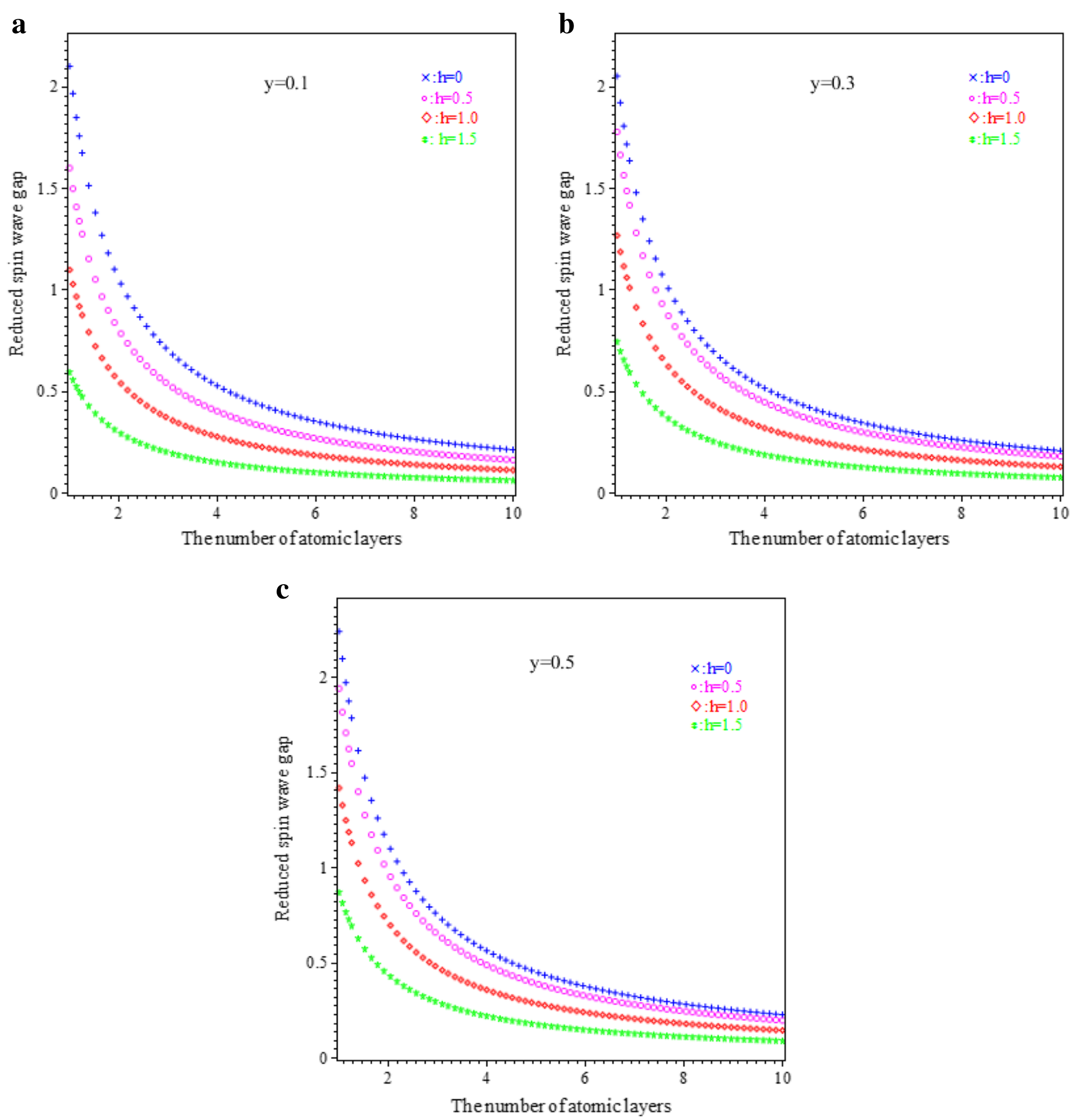

Fig. 3 Reduced spin wave gap as a function of the number of atomic layers for different reduced external magnetic fields and values of y: **a** $y = 0.1$, **b** $y = 0.3$ and **c** $y = 0.5$

for magnetic anisotropies are usually interpreted in terms of competition between surface and volume attributions and the magnetic anisotropy. However, the origins of these anisotropy contributions are not very clear so far. Experimental results on Co films covered with 2 ML Cu indicate that the out-of-plane anisotropy is drastically converted, ruling out a standard Neel-type surface anisotropy mechanism which is caused by the broken translational symmetry at the surface along its normal [26, 27].

For ultrathin films, in which the thickness of the film is smaller than the exchange length, apart from a perpendicular surface anisotropy contribution, an in-plane anisotropy of second-order symmetry could be separated into a thickness-independent volume term K_i^V and a surface term k_i in which $i = o, p$ for perpendicular and in-plane anisotropy respectively. It can be interesting to look at the behavior of the spin wave gap for the different values of y. In (6), we have used $n = \frac{L}{a_o}$ which indicates the number of atomic layers, where a_o is the lattice spacing. In Fig. 3a–c, we plot the calculated spin wave gap as a function of the number of atomic layers for various values of y and for the small values of the critical fields $h \leq 2$. Obviously, when y is increased, the calculated spin wave gap at zero temperature is found to gradually increase for films consisting of two or three atomic layers and, as h is increased, the spin wave gaps of almost the same thickness dependence are characterized by their typical $\frac{1}{L^2}$ behavior [7]. In this condition, the in-plane anisotropy greatly influences the spin wave gap.

In Fig. 4, the spin wave gap is plotted as a function of the applied magnetic field for three different atomic layers as an experimental value of $y = 0.5$. Obviously, at small values of h, the shape anisotropy is large compared to the influence of the in-plane anisotropy. This leads to a decrease of the lowest spin wave gap with increasing h, which is similar to the behavior observed in nanocrystalline CoFeB films with growth-induced anisotropy [13]. At larger values of h, the in-plane anisotropy dominates and the spin wave gaps increase as we expected.

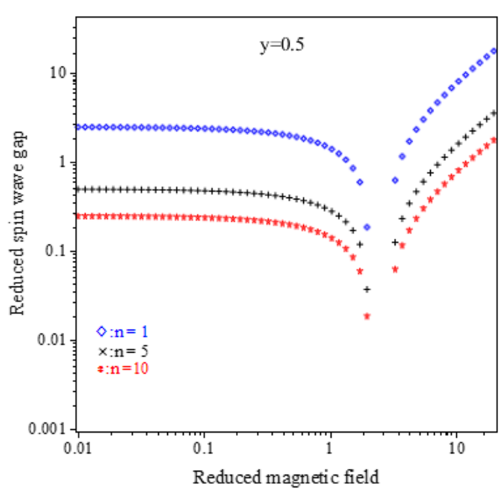

Fig. 4 Reduced spin wave gap as a function of the reduced applied magnetic field for three atomic layers. The value of y was held constant at 0.5

In conclusion, we have developed a quantum theory describing the spin wave energy gap in single-layer and thin magnetic films that include both out-of-plane and in-plane anisotropies for a general direction of the applied magnetic field, H. The most important property of the calculated equations presents a nonzero spin wave gap at zero field. Such a gap is strongly affected by anisotropies and external fields. When the intensity of the in-plane anisotropy is increased, a qualitatively different behavior is found depending on the intensity of the applied magnetic field. The effects of film thickness are also examined. The spin wave gap decreases with an increase of the number of atomic layers. Our result confirms that the measured anisotropies are more crucial in determining the spin wave gap and so validates the presented theoretical model. A comparison of our results with the recent measurements of the field dependence of the spin wave gaps shows a beautiful agreement between theory and experiment.

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