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## Anisotropy effects on the spin wave gap of two dimensional magnets at zero temperature



B. Kaplan\*, R. Kaplan

Department of Secondary Science and Mathematics Education, University of Mersin, Yenisehir Campus, 33169 Mersin, Turkey

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## ABSTRACT

The principal purpose of this paper is to report an estimate of spin-wave gap of two-dimensional magnets taking into account both the anisotropy and the Zeeman energy sufficiently large to dominate over the dipolar interaction. The spin-wave gap is calculated for a magnetic field which is perpendicular to the plane at zero temperature. The results are discussed in connection with experimental data reported for epitaxial Fe-deficient yttrium garnet (YIG) films pulsed laser deposited onto the different faces of  $Gd_3Ga_5O_{12}$  single crystal.

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## 1. Introduction

Magnetic properties of magnetic materials in which the film thickness varies from two to three dozens of unit cells are a subject of growing interest in recent years both experimentally and theoretically. Magnetic anisotropy plays a key role in the physics of ultrathin magnetic films. It affects the frequencies of spin-wave excitations, the nature of the domain walls, and the magnetization reversal process. The magnetic anisotropy originates from the magnetic dipolar interaction (shape anisotropy) and the magnetocrystalline anisotropy due to the spin-orbit interaction. The magnetic dipolar interaction always chooses in plane magnetization in thin films but the spin-orbit interaction shows clearly much more complicated behaviors. In addition, physically the experimental data for magnetic anisotropies are usually interpreted in terms of competition between surface and volume attributions and the magnetic anisotropy. Nonetheless, there are difficulties for understanding the direction of magnetization in such a way that the separation of these two effects is not physically clear in ultrathin films.

The inclusion of the magnetic anisotropy causes an energy gap in the spin-wave spectrum. Such a gap acts as an energy barrier to the excitation of long wavelength spin waves, thus allowing a finite order parameter at finite temperatures. For the case of a uniaxial anisotropy favoring in-plane magnetization it has been proposed that an effective gap arises due to the long range character of the dipole interaction [1,2]. Bruno [1] finds for the case of dipolar interactions only and the role of in-plane magnetocrystalline anisotropy is not included in this calculation. The origin of the observed thermal stability of the magnetization [3,4] is associated with the spin-wave gap induced by the in-plane

anisotropy field [5]. The effect of an in-plane anisotropy is dominant in stabilizing the in-plane magnetization of a two-dimensional ferromagnetic film when compared to the effect of dipolar interactions alone [6].

Experimentally, a size-effect for the temperature of magnetic transition appears in  $MnF_2$  epitaxial films with an orthorhombic crystal structure in thick films (120–1250 nm) which is contributed to the low anisotropy energy inferred from the spin-wave gap [7]. Investigations of the nature of magnetic anisotropy for iron garnet films grown by rf-sputtering [8,9] and the pulsed laser deposition technique (PLD) [10,11] are of great interest because of remarkable magneto-optic properties. This material is very interesting for applications involving significant magneto-optic and non-reciprocal effects [12]. Popova et al. [12] showed that the production of the magnetophotonic crystal structure lowers the magnetocrystalline and uniaxial in-plane anisotropies and induces a partial out-of-plane magnetization. Recently, an unusual magnetic anisotropy has been observed in epitaxial Fe-deficient yttrium iron garnet (YIG) films pulsed laser deposited onto the (111) and (001) faces of  $Gd_3Ga_5O_{12}$  single crystal and also explained the effects of reduced cubic and strong negative growth induced uniaxial magnetic anisotropy for these films [13].

It can be of interest to look at the spin-wave gap of two-dimensional materials as a function of magnetic field which is applied perpendicular to the film plane at zero temperature.

## 2. Calculation of spin wave gap

As a starting point, we consider a film in the  $(x,y)$  plane and assume that the total Hamiltonian can be expressed generally as follows:

$$H = H_Z + H_{exc} + H_a \quad (1)$$

\* Corresponding author. Tel.: +90 324 3412815x2032; fax: +90 324 3412823.  
E-mail address: [bengukaplan@yahoo.com](mailto:bengukaplan@yahoo.com) (B. Kaplan).

Here the Hamiltonian  $H$  contains the Zeeman term, an exchange term, and an anisotropy term respectively. It is assumed that all magnetic atoms have the same spin quantum number  $S$ . The first term of Eq. (1) represents the Zeeman energy of the spins in a field of magnitude  $h$ , which throughout we suppose is directed along the  $z$  axis. The second term represents the exchange energy; the proof that it is proportional to the scalar product of the two spin vectors is quite familiar [14] and is omitted here. The final term in Eq. (1) describes the magnetic anisotropy energy which has the following form:

$$H_a = \sum_n [K_2 S_z^2(n) - K_4 S_z^2(n) (S_x^2(n) + S_y^2(n)) - K'_2 S_y^2(n) - K'_4 S_x^2(n) S_y^2(n)] \quad (2)$$

so that this film as a cubic material, normal parallel to  $\hat{z}$ , is used to obtain the above equation. The out-of-plane anisotropy is expressed in terms of a second-order uniaxial anisotropy  $K_2$  which includes the shape anisotropy term and the modified fourth-order cubic anisotropy  $K_4$ . The in-plane anisotropy term is given in terms of a uniaxial term  $K'_2$  which breaks the fourfold symmetry and a modified cubic anisotropy constant  $K'_4$ , and all of them depend on the film thickness  $L$ . The sign convention contained in Eq. (2) implies that positive (negative) values of anisotropy constants favor the magnetization lying perpendicular to the film plane (in the plane).

The dipolar interactions contribute to the total anisotropy  $K_2$  and also modify the form of the spin-wave spectrum, thus removing the logarithmic divergence in the magnetization due to spin waves of small  $k$  [15] which denotes the spin-wave wave vector. We note that there are two differences among the effect of anisotropy  $K'_4$ , an easy plane film and a cubic crystal. For the bulk crystal where  $K_2 = 0$  the spin-wave gap is proportional to  $K'_4$  and since in the three dimensional case the integrated spin deviation converges to  $K'_4 = 0$ , the effect of  $K'_4$  is usually neglected.

We start with the assumption that the equilibrium orientation of all the spins is along the  $z$  axis. To examine the spin wave gap at zero temperature we first use the Holstein–Primakoff transformation [16]. Corresponding to our choice of coordinate axes, the components of a spin vector  $S(n)$  are represented in terms of boson creation and annihilation operators  $a_n^+$  and  $a_n$  by the following expressions:

$$S^z(n) = S - a_n^+ a_n, \quad S^x(n) = \frac{\sqrt{2S}}{2} (a_n + a_n^+), \quad S^y(n) = \frac{\sqrt{2S}}{2i} (a_n - a_n^+).$$

The exchange interaction has the largest energy in the monolayer films. For large values of  $k$ ,  $\varepsilon_k$  are given by

$$\varepsilon_k \cong Dk^2 \quad (3)$$

where  $\varepsilon_k$  arises from the exchange forces and  $D \cong 2JSa^2$ , in which  $J$  is the Heisenberg exchange energy,  $S$  is the spin operator and  $a$  is the lattice parameter. The total spin-wave Hamiltonian can be written in the following form [1]:

$$H = \sum \left[ A_k a_k^+ a_k + \frac{B_k}{2} (a_k^+ a_{-k}^+ + a_k a_{-k}) \right] \quad (4)$$

where  $A_k = \varepsilon_k + h_o + \alpha$  and  $B_k = \beta$ .

Here  $\alpha$  and  $\beta$  arise from the anisotropy energy and are independent of  $k$ ,  $h_o$  is the external magnetic field, and the Zeeman interaction factors  $g\mu_B$  (gyromagnetic ratio and Bohr magneton) have been absorbed in the definition of  $h_o$ . The spin-wave energy is given by the following expression:

$$\omega_k^2 = |A_k|^2 - |B_k|^2. \quad (5)$$

This is of the same form as discussed by Bruno [1] for the dipolar case at zero magnetic field. For our Hamiltonian we find

(with  $S=1$ ) the following expression:

$$\omega_k^2 = (\varepsilon_k + h_o - 2K_2 - 2K_4)(\varepsilon_k + h_o - 2K_2 - 2K_4 - 2K'_2). \quad (6)$$

This gives a spin-wave gap at  $k=0$ ,

$$\omega_0^2 = (h_o - 2K_2 - 2K_4)(h_o - 2K_2 - 2K_4 - 2K'_2) \quad (7)$$

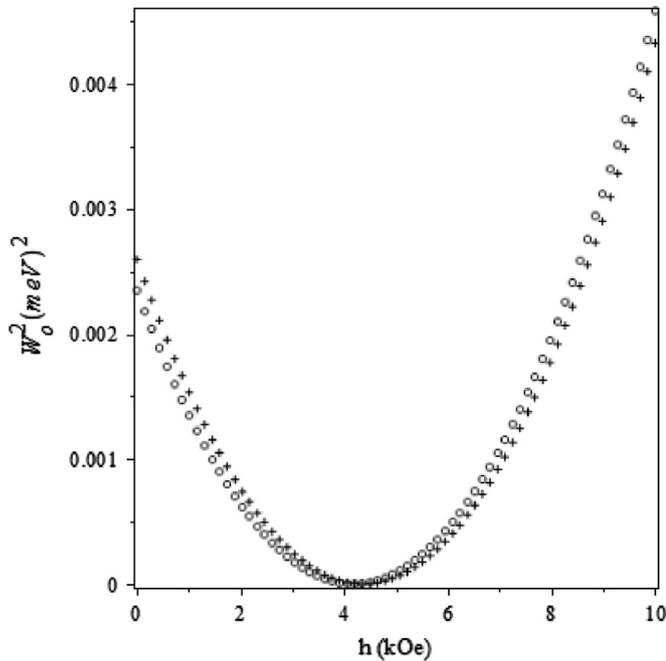
which is not zero when the in-plane anisotropy  $K'_2$  vanishes. We note that this effect arises from the cubic anisotropy allowed term  $K_4$  and that the symmetry breaking term  $K'_2$  only adds to the magnitude of the gap and can be easily eliminated. It is useful to introduce the anisotropic fields  $K_2 = g\mu_B H_2$  and  $K_4 = g\mu_B H_4$ , and we obtain the following expression:

$$\omega_0^2 = (g\mu_B)^2 (h - 2H_2 - 2H_4)^2. \quad (8)$$

Manuilov and Grishin [13] have measured the twofold and fourfold anisotropy fields and also gyromagnetic  $g$ -factor as a function of three different orientations of external magnetic field: perpendicular to the film plane  $h//[001]$  and two in-plane directions  $h//[100]$  and  $h//[110]$ . In the computation, in perpendicular magnetic field ( $h \leq 10^4$  Oe),

$H_2 = -2029$  Oe,  $H_4 = -57$  Oe, and  $g_{//} = 2.004$  for  $L = 140$  nm; and  $H_2 = -2152$  Oe,  $H_4 = -31$  Oe, and  $g_{//} = 2.014$  for  $L = 200$  nm films. Although Manuilov and Grishin [13] have observed twofold and fourfold anisotropy fields in some of their materials, they did not report any measured uniaxial term  $H'_2$  which breaks the fourfold symmetry. This contribution only adds to the magnitude of the gap, and can be easily eliminated. In perpendicular magnetic field in Ref. [13], the ferromagnetic resonance demonstrates an occurrence of soft mode. As seen from Eq. (8), the spin-wave gap tends to zero in the magnetic field  $h = 2H_2 + 2H_4$ . Manuilov and Grishin [13] show that the uniform perpendicular magnetized state becomes unstable with respect to the nucleation of magnetic domains with in-plane components of magnetization tended along some different directions. They detected this reorientation phase transition at very low frequencies. Their experimental data are well suited for our theoretical results. Fig. 1 shows a plot of our expression for  $\omega_0^2$  as a function of  $h$ . We calculate the spin-wave gaps in the Kelvin range as 0.710 K (5.254 kOe) and 0.820 K (6.068 kOe) for  $L = 140$  nm and  $L = 200$  nm respectively. Manuilov and Grishin [13] have reported the resonance fields between 5 and 5.5 kOe in their films. In comparison, a good coincidence is obtained between our and their results. As seen in Fig. 1 the important property is that both curves have a fast minimum at the critical field of about 4.277 kOe, and thus the spin-waves indicate distinct behavior for the external field being less than or greater than this critical value. For the external fields larger than the critical value, the spins are perpendicular to the plane of film, and spin-wave gaps increase monotonically which is mainly due to the Zeeman energy term. On the other hand, for the fields smaller than the critical value, the spins are canted slightly according to the film plane. The addition of the Zeeman energy to the spin-wave gap is overwhelmed by the demagnetizing energy and the curves decrease slowly with increasing applied field. Note that the spin-wave gap is vanished at the critical value of 4.277 kOe. This is only satisfied if the applied field equals the out-of-plane anisotropies.

A similar behavior given above has also been observed in ferromagnetic nanowires in the presence of dipolar interactions and the Heisenberg-exchange interactions between nearest neighbors [17]. They have also reported that if the applied field is perpendicular to the ferromagnetic nanowire axis, there exists a canting of the net spin reorientation which is away from the axis, and the magnetization is spatially nonuniform owing to dipolar interactions and that typically there are two phases and two

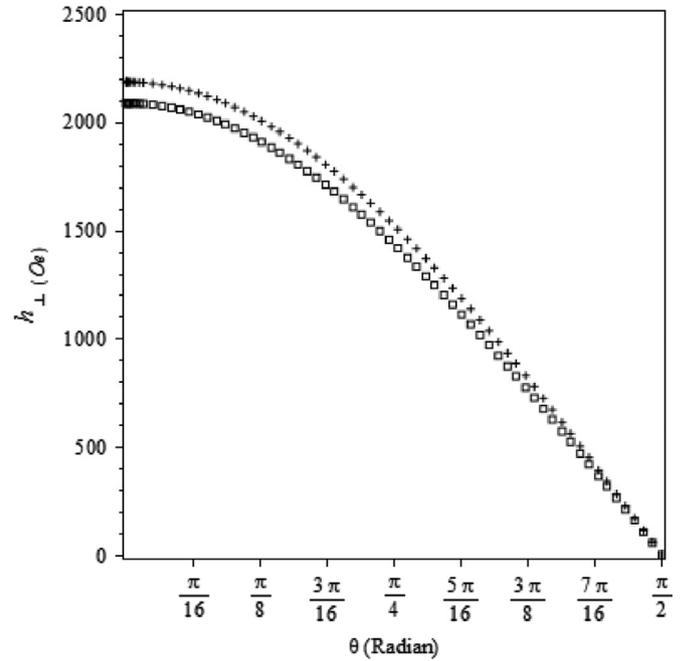


**Fig. 1.** Magnetic field dependence of the energy for  $L = 140$  nm (circle lines) and  $L = 200$  nm (cross lines).

distinct regimes of spin-wave behavior, corresponding to the applied field which is being less than or greater than a critical value.

We now consider the polar coordinates  $\theta$  and  $\varphi$ , where  $\theta$  measures the angle between the  $z$  axis and vector  $S$  and the azimuthal angle  $\varphi$  measures the angle between the projection of  $S$  on the  $(x, y)$  plane and the  $x$  axis. The total anisotropy energy is obtained from the Hamiltonian given in Eq. (2). The equilibrium configuration must be determined from the condition of the minimum of the total anisotropy energy. Fig. 2 shows the calculated perpendicular magnetic field variation as a function of the angle  $\theta$  in films with two different thicknesses according to the experimental data given in Ref. [13]. As can be seen, the applied perpendicular magnetic field decreases with decreasing film thickness and also the precession of magnetic spins takes place in the applied field due to different magnetic anisotropies and interactions in the magnetic film. Popova et al. [12] reported that the applied external field must be less than 2 kOe in order to ensure the out-of-plane saturation of magnetization for optimum device operation. Although our calculation is little larger than their result (2 kOe), it is very close and gives support to it.

In conclusion, our result confirms that the measured anisotropies are more crucial in determining the spin wave gap and so validates the presented theoretical model. The spin-wave energy gap does not vary



**Fig. 2.** The magnetic field applied perpendicularly to the film plane as the rotation angle  $\theta$  for films with various thicknesses ( $L = 140$  nm (box lines) and  $L = 200$  nm (cross lines)).

linearly with the in- and out-of-plane anisotropies but is magnified because it is the geometric mean of these anisotropy energies which is very important.

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