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ORIGINAL PAPER

In-Plane Anisotropy Effect to the Spin-Wave Gap in Ultrathin Ferromagnetic Films at Finite Temperatures

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Abstract

In this paper, we investigate the calculated thickness- and temperature-dependent magnetization given in terms of the spin-wave gap alone for two-dimensional ultrathin ferromagnetic films with anisotropy and Zeeman energy sufficiently large to dominate over the dipolar interaction. The spin-wave gap was calculated for a magnetic field which is perpendicular to the plane. The calculated equations present a nonzero spin-wave gap at zero magnetic field which is strongly affected by anisotropies. The temperature-dependent magnetization strongly increases with decreasing thickness of the insulating spacer layer at the saturation field and at room temperature. We reported that the in-plane anisotropy strongly depends on the insulating spacer layer thickness and saturation field and overcomes the fourfold anisotropy in the spacer layers with thickness of below 0.8 nm. The results were also discussed in connection with experimental data given in $Co_{0.9}Fe_{0.1}/MgO/C_{0.9}Fe_{0.1}$ films.

Keywords Magnetic anisotropy · Spin-wave gap · Temperature- and thickness-dependent magnetization

1 Introduction

One of the actively investigated subjects is the magnetic anisotropy which is closely related to the utilization of direction of magnetization in thin films and two-dimensional magnetic materials grown on magnetic and/or nonmagnetic substrates due to possible applications in ultrahigh magnetic and magnetooptic data storage devices and sensors [1–3]. In many cases, the reorientation transitions of the magnetization are driven by variations of the relative film thicknesses, temperature and external magnetic field [4–12].

Quite recently, the reorientation transitions have been observed to be induced by very thin ferromagnetic/antiferromagnetic system [13] and ferromagnetic metal/insulator/ferromagnetic metal magnetic tunnelling junctions [14] with perpendicular magnetic anisotropy which have great advantages over the in-plane ones at room temperature. The magnetization at room temperature can be significantly reduced from the ground-state value due to thermally excited spin waves. A stronger temperature dependence of the magnetization occurs in ultrathin films in comparison with the bulk. Lasek et al. [14] have observed an influence of the thickness of the insulating spacer layer, t_{MgO}, on the magnetic anisotropy, especially near the spin reorientation transition, in *ferromagnetic metal/insulator/ferromagnetic metal* structures at room temperature. They reported that when the thickness of the insulator of MgO is more decreased, the contribution of in-plane anisotropy energy components increases and the coexistence of vertical and in-plane easy directions arise. The magnitudes of the magnetic anisotropy constants were determined from ferromagnetic resonance and dc magnetometry measurements. The origin of magnetic anisotropy evolution has been attributed to the presence of an interlayer exchange coupling between ferromagnetic layers through the thin MgO film.

In a previous work [15], we calculated the spin-wave gap as a function of the applied magnetic field which is perpendicular to the film plane and the angular dependence of magnetization reversal in a single-layer thin magnetic film that includes the strong perpendicular anisotropy at the absolute zero of temperature. We concluded that the in-plane anisotropy contribution influences the spin-wave gap of ultrathin films with small thickness at zero temperature. In this study, it can be of interest to look at the thermal

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variation of the magnetization as functions of the film thickness, spin-wave gap, exchange temperature, applied field and field direction.

2 Theoretical Model

We first consider a film which is L_z layers thick which is the number of atomic layers (ML) and extends over $L_x L_y$ cells in plane. To evaluate the spin-wave energy gap, one has to start with a description of the system's magnetic energy. The total magnetic energy density in an external magnetic field can be written as

$$E = -M_S H \cos(\theta - \theta_H) + E_A(\theta, \varphi) \tag{1}$$

where the first term represents the Zeeman energy of the magnetic layer with M (magnetization) oriented at an angle θ from its normal as the result of the applied magnetic field (H) at the angle θ_H with respect to the normal to the layer's surface and also M_S is the saturation magnetization. The second term, $E_A(\theta, \varphi)$, is the magnetic anisotropy energy which contributes to the perpendicular interface anisotropy as well as the uniaxial in-plane one

$$E_A(\theta, \varphi) = -K_1(\mathbf{s}.\mathbf{e_z})^2 - K_2(\mathbf{s}.\mathbf{e_z})^4 + K_3(\mathbf{s}.\mathbf{e_y})^2 \tag{2}$$

where the out-of-plane anisotropy is expressed in terms of the second-order uniaxial anisotropy (K_1) which includes volume, surface and shape anisotropies and the fourth-order anisotropy (K_2). The in-plane anisotropy is given in terms of a uniaxial anisotropy (K_3) which breaks the fourfold symmetry. All of the anisotropy constants depend on the thickness of the films, the temperature and the nonmagnetic spacer layers. In (2), $\mathbf{e_y}$ and $\mathbf{e_z}$ are the unit vectors defining the anisotropy axes, respectively; $s = f(\theta, \varphi)$, $\| s \| = 1$ and φ defines the in-plane orientations. The sign convention in (2) implies that positive (negative) values of anisotropy constants favour the magnetization lying perpendicular to the film plane (in the plane). The spin-wave energy gap (ω_0) corresponds to a temperature (T_g) which is given by the following expression [17, 23]:

$$\omega_0 = k_B T_g = \left[\frac{\partial^2 E}{\partial \theta^2}\Big|_{\theta_0,\varphi_0} \frac{\partial^2 E}{\partial \varphi^2}\Big|_{\theta_0,\varphi_0} \right]^{1/2} \tag{3}$$

where k_B indicates the Boltzmann constant which is given in units of 1.3807×10^{-16} erg/K. In (3), θ_0 and φ_0 are the initial values of θ and φ, which minimize E for some value of the applied field and field direction and are evaluated from

$$\left.\frac{\partial E}{\partial \theta}\right|_{\theta_0,\varphi_0} = \left.\frac{\partial E}{\partial \varphi}\right|_{\theta_0,\varphi_0} = 0.$$

Lasek et al. [14] have measured the second- and fourth-order uniaxial anisotropies in some of their materials at room temperature as a function of the thin insulating spacer layer. Experimentally, the saturation magnetic field strength is 8 kOe in the perpendicular orientation. The values they found are as follows: $K_1 \simeq \pm 2.5 \times 10^5 \text{erg/cm}^3$ and $K_2 \simeq 1.9 \times 10^5 \text{erg/cm}^3$ for $t_{MgO} = 0.8$ nm and $K_1 \simeq 7.5 \times 10^5 \text{erg/cm}^3$ and $K_2 \simeq 1.7 \times 10^5 \text{erg/cm}^3$ for $t_{MgO} = 1.0$ nm of insulating spacer layers and for $L(Co_{0.9}Fe_{0.1}) = 1.4$ nm film. However, they have not measured any values for in-plane term K_3 which breaks the fourfold symmetry. This attribution only adds to the magnitude of the spin-wave gap; however, it is important for our calculations. Our theoretical results are well suited for their experimental data given [14], except for in-plane anisotropy. We have used the above experimental values, such as $H \geq 8$ kOe and the dimensionless parameters $k = \frac{K_2}{K_1} = \pm 0.76$ for 0.8 nm and $k = 0.23$ for 1.0 nm. Figure 1a, b shows the spin-wave gaps as a function of the dimensionless parameter $\eta = \frac{K_3}{K_1}$ for only positive k value at zero and 8 kOe magnetic fields for two insulating spacer layers. At zero magnetic field, the gap is essentially determined by the magnetic anisotropy as we have expected. As can be seen, the variation in energy gaps is due to the in-plane anisotropy presented in the film. One striking feature of these plots is that the amplitude of the energy gap variation increases gradually for the higher-order t_{MgO} which is not linear for both cases of perpendicular easy-axis and easy-plane anisotropy [16, 17]. Lasek et al. [14] observed that while the magnetic anisotropy of the top layer does not change its character upon variation of the t_{MgO}, the uniaxial out-of-plane magnetic anisotropy of the bottom, one undergoes a spin reorientation transition at the thickness of MgO of about 0.8 nm, switching to the regime where the coexistence of in-plane and out-of-plane magnetization alignments is observed. Figure 2a, b shows the calculated spin-wave gaps as a function of the applied magnetic field with respect to the [001] axis for two different insulating spacer layers for various values of η. Obviously, the curves have a sharp minimum at the zero field, but they do not vanish in which the three lines are very close each other in Fig. 2a and this situation is different in Fig. 2b. The spin waves show distinct behaviour for the applied field being less than or greater than the zero field. We interpret this as a consequence of non-uniform magnetization. Experimentally [14], the mixed anisotropy case present for larger t_{MgO} transforms to a case where both ferromagnetic layers exhibit the easy-plane magnetic anisotropy. 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. Furthermore, 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. On the other hand, for $H > 0$, the in-plane anisotropy dominates and spin-wave gaps increase

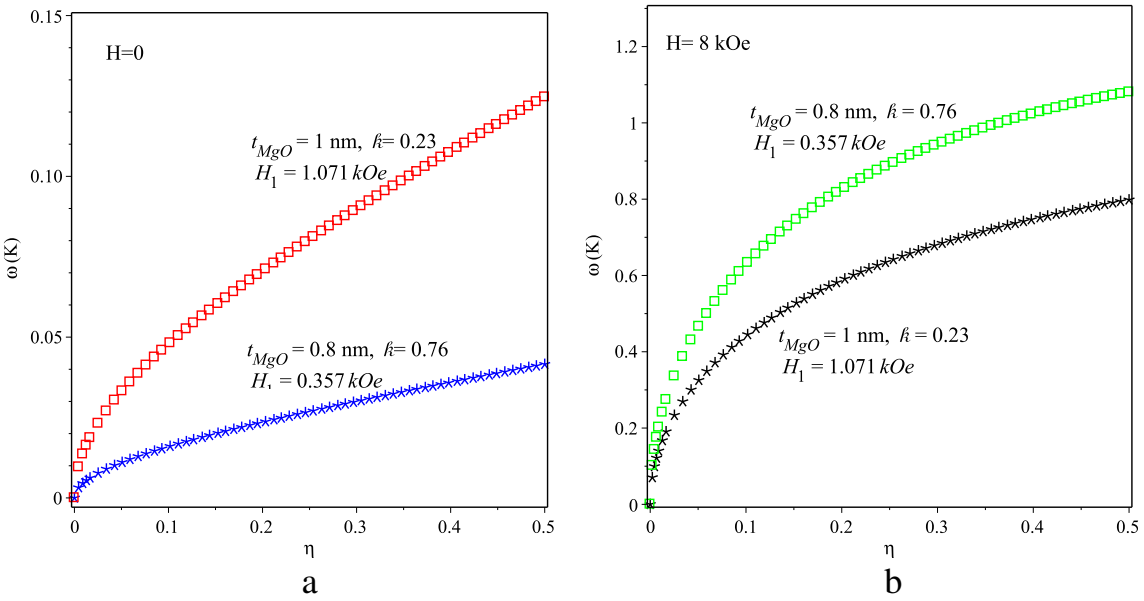

Fig. 1 Spin-wave gap as a function of η for $H = 0$ (**a**) and $H = 8$ kOe (**b**)

uniformly. This indicates that the film is uniformly magnetized. A qualitative similar behaviour has been observed in ferromagnetic films with different anisotropy contributions [18, 19]. However, if we use the negative value of k for $t_{\mathrm{MgO}} = 0.8$ nm in Fig. 2a, the ω increases very sharply with increasing H values. In this case, the curves vanish at zero field which are not shown on the figure.

We next concentrate to achieve a better understanding of the thickness- and temperature-dependent magnetization of ultrathin magnetic films which is a subject of strong current interest [9, 10, 13] with different kinds of magnetic anisotropies. The magnetization of two-dimensional magnets must decrease linearly with the temperature [20, 21]. On the other hand, some other workers presumed that two-dimensional magnetic materials with a perpendicular easy-axis must represent a linear temperature dependence of the magnetization, in contrast to two-dimensional magnets with easy-plane anisotropy [22]. The temperature-dependent magnetization is given by $T \ln(T)$, which is clearly not linear for both the cases of perpendicular

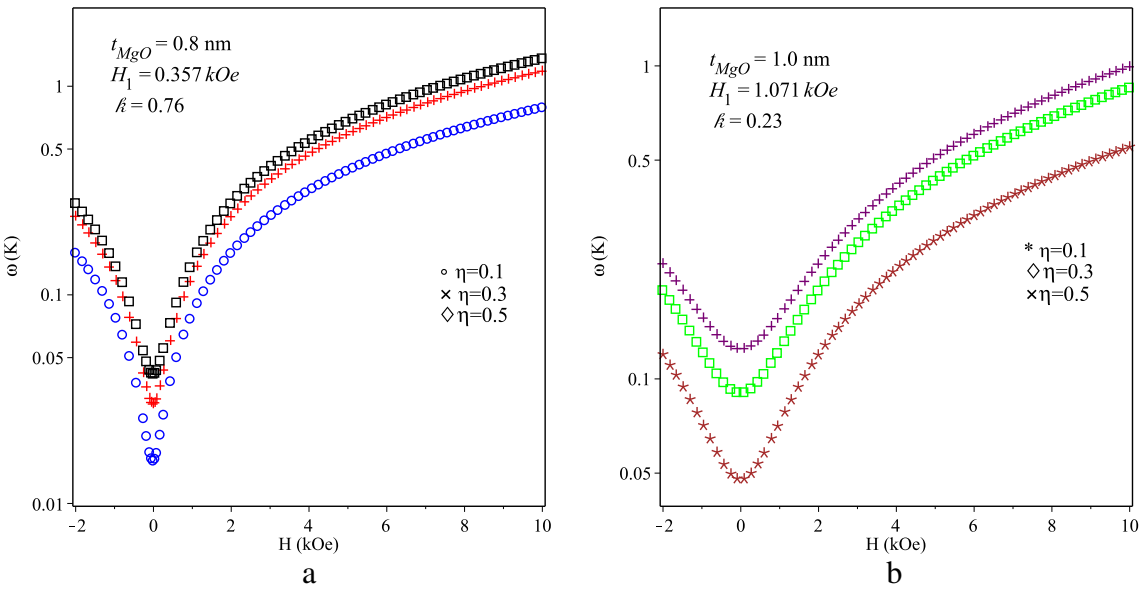

Fig. 2 Field dependence of the spin-wave gap for different values of η for $t_{\mathrm{MgO}} = 0.8$ nm (**a**) and $t_{\mathrm{MgO}} = 1.0$ nm (**b**)

easy-axis and easy-plane anisotropy [16, 17]. The magnetic anisotropy leads to a spin-wave gap (T_g) from which the thickness- and temperature-dependent magnetization can be calculated within the framework of non-interacting spin-wave theory. The normalized change in the magnetization of a film of L_z layers thick as a function of temperature (T), the applied magnetic field (H) and field direction (θ_H) is given by [17, 23]

$$\frac{\Delta M(T,H,\theta_H)}{M_S}=\frac{T}{T_{ex}L_z}\ln\left[\frac{T}{T_g(H,\theta_H)}\right] \quad (4)$$

where T_{ex} is determined by the atomic spin and the strength of the exchange interaction and is given by $T_{ex}=8\pi s\mu_B H_{ex}k_B^{-1}$, which is called the exchange temperature of the film, so that μ_B is given in units of 6.72×10^{-2}K/kOe which is well known as Bohr magneton, and H_{ex} denotes the effective field. In (4), $T_g=\omega_0/k_B$ is the spin-wave energy gap in Kelvin range as a function of the applied magnetic field and its direction. The expression for T_g can now be used in conjunction with (4) to calculate the magnetization deviation. Thus, the resulting expression for T_g depends on the magnetic anisotropy strengths of the film, the number of atomic layers, the exchange temperature and the strength and orientation of applied magnetic field. The spin-wave gaps (T_g) given in Kelvin units are also listed in Table 1 at $H=0$ and at $H=8$ kOe. By using experimental data, we obtained the theoretical values of the spin-wave gap temperatures given in Table 1 from Fig. 2a, b. As can be seen from Table 1, the larger values of the spin-wave gaps are obtained for $\eta=0.3$ and 0.5 which are very close to each other at the saturation field for $t_{MgO}=0.8$ nm and $t_{MgO}=1$ nm. Experimentally, the magnetic moments on the surface are progressively affected by the interlayer interactions and thus the surface anisotropy energy, and therefore, the magnetic anisotropy energy changes [14]. It is interesting in this context to note that the largest spin-wave gaps suggest that the interface magnetism is likely to play the most marked role [23]. Clearly more accurate investigations of the ferromagnetic metal/insulator/ferromagnetic metal structures are required in the future to clarify the role of the interface of the insulating spacer layers in influencing the temperature-dependent magnetization.

Figure 3 shows the temperature-dependent magnetization as a function of the exchange temperature for two different insulating spacer layers for only $\eta=0.3$ [15] for one ML film at zero and saturation fields of 8 kOe at room temperature. It is interesting to note that the magnetization was estimated in zero and saturation field as $M\sim0.5M_S$ and $M\sim0.6M_S$, respectively, at $T_{ex}\sim3400$ K, which corresponds to the bulk Co [16]. Obviously, the magnetization is larger for 8 kOe than that of zero field as expected. Lasek et al. [14] observed that the sample with $t_{MgO}=1.0$ nm exhibits a two-step character, corresponding to easy-axis and hard-axis magnetization processes at room temperature. The sloppy part of the M starts from $M\approx0.5M_S$, achieved at 8 kOe. Besides, on lowering t_{MgO}, the system is losing the initial easy-axis behaviour, indicating a hard-axis character. All measurements were performed in the perpendicular orientation of H. The anisotropy constants provide the conditions for the coexistence of the two easy directions of magnetization, which the first one is along the stable z-axis and the other one is on the metastable sample plane, if the thickness of the insulating spacer layer decreases below 0.8 nm. If the thickness of the insulating spacer layer is below 0.8 nm, the above two easy directions replaced each other. In other words, the stable z-axis becomes metastable and the metastable sample plane becomes stable, which are in agreement with our results. The observed magnetic anisotropy progression for the $t_{MgO}<0.8$ nm can be referred to the presence of an interlayer exchange coupling between their ferromagnetic $Co_{0.9}Fe_{0.1}$ layers through the thin MgO film. The addition of in-plane anisotropy energy components increases, and the coexistence of vertical and in-plane easy directions appears, when the thickness of the insulator of MgO is further decreased [14].

It can be of interest to look at the temperature dependence of magnetization at low temperature in order to demonstrate how the influence of the thickness of the insulating spacer layer varies with the influence of the zero and saturation field. These are shown in Fig. 4 for the one ML film in which the two different insulating spacer layer thicknesses are 0.8 and 1 nm for $\eta=0.3$ at 5 K. As it is shown, the magnetization is almost close to a constant value of $M\sim0.99M_S$ above about $T_{ex}>2000$K. This corresponds to the bulk Fe for 2300 K [23] at zero field. At a saturation field of 8 kOe, however, the insulating spacer layer thickness of 0.8 nm has a larger value than that of 1.0 nm. Obviously,

Table 1 The calculated spin-wave gap temperatures deduced from fits to the experimental data

η	T_g(K); $t_{MgO}=0.8$ nm		T_g(K); $t_{MgO}=1$ nm	
	$H=0$	$H=8$kOe	$H=0$	$H=8$kOe
0.1	0.01	0.62	0.05	0.44
0.3	0.03	0.96	0.09	0.66
0.5	0.04	1.06	0.12	0.79

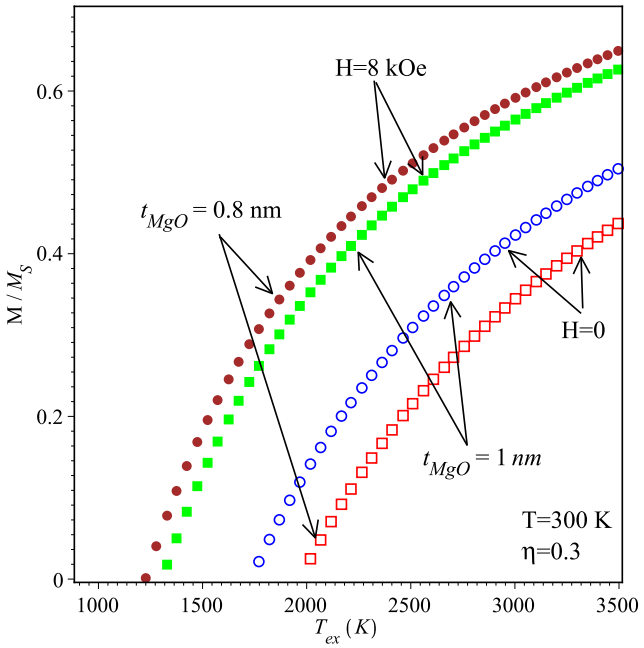

Fig. 3 The normalized magnetization as a function of exchange temperature at room temperature (300 K) for $H = 0$ and $H = 8$ kOe

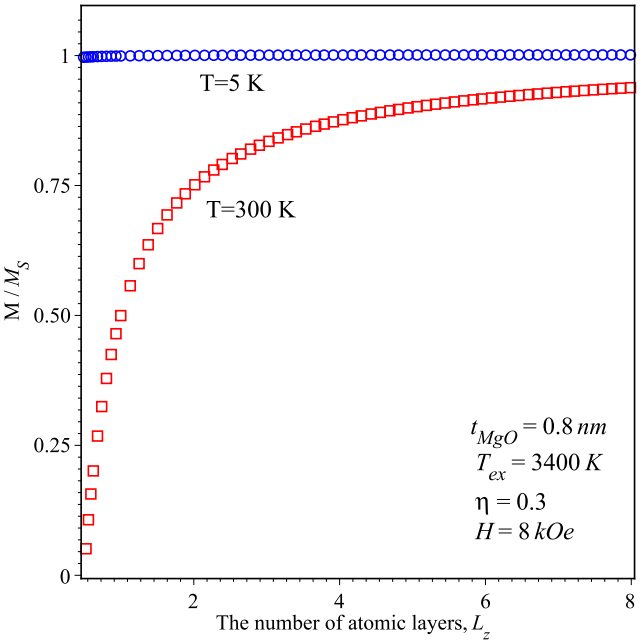

Fig. 5 The normalized magnetization with respect to the number of atomic layers at 5 and 300 K

our results indicate the fact that the in-plane anisotropy is strongly thick and saturation field dependent and overcomes the fourfold anisotropy in the spacer layers with a thickness of below 0.8 nm.

Figure 5 shows the normalized magnetization as a function of the number of atomic layers, L_z at two different temperatures of 5 and 300 K. As seen, at 300 K, the normalized magnetization decreases with the decrease of the number of atomic layers and falls sharply for $L_z \leq 2$. Similar results were experimentally observed for Fe/Ag [8] and Fe/Mn [12] multilayer films by using Bloch's spin-wave expression at room temperature. However, at 5 K, it does not change significantly and remains almost constant. This behaviour can be attributed to the finite size effect [12].

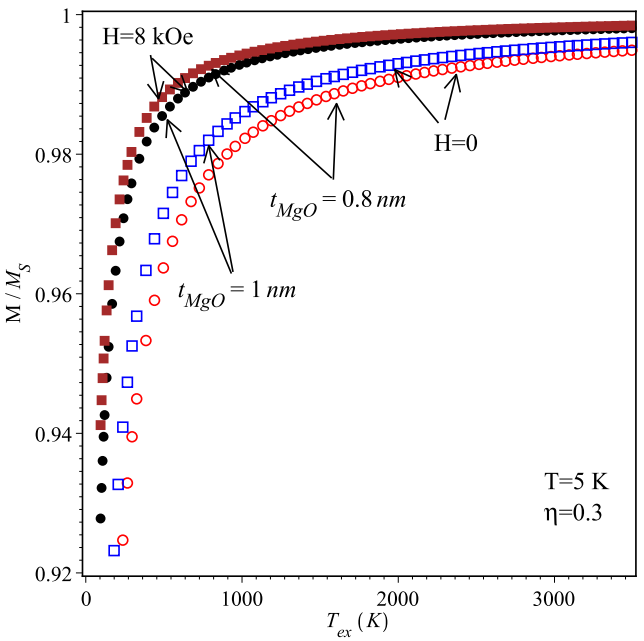

Fig. 4 The normalized magnetization vs the exchange temperature at low temperature (5 K) for $H = 0$ and $H = 8$ kOe

In conclusion, our calculations confirm that the in-plane anisotropy contribution influences the spin-wave gaps of ultrathin films with small thickness. The in-plane anisotropy should dominate in determining the spin-wave gap by over an order of magnitude. It is important to rule out the possible effect of an applied field in determining the spin-wave gap which validates the presented models. We also discussed the temperature-dependent magnetization as a function of the number of atomic layers, the exchange temperature and the strength and orientation of the applied magnetic field. The magnetization strongly increases with decreasing thickness of the insulating spacer layer at saturation field at low temperature as well as at room temperature. We can reported that our results present that the recent measurements of the second- and fourth-order anisotropies at room and low temperatures show a beautiful agreement between theory and experiment.

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