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Influence of magnetic surface anisotropy on the dynamic properties in ferromagnetic thin films

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Abstract

A Green's function technique is applied for the Heisenberg model to study the influence of the magnetic surface single-ion anisotropy on the spin wave spectrum including damping effects in ferromagnetic thin films. It is shown that the magnetic surface anisotropy strongly affects the thickness dependence of different quantities: for strong surface anisotropy, the magnetization, the spin-wave energy and the phase transition temperature T_C are larger in thin films than in the bulk, whereas the opposite is true for small surface anisotropy. The magnetic surface anisotropy enhances the spin-wave damping. There is some competition between the surface single-ion anisotropy and the surface exchange interaction. The effect of an external magnetic field is discussed, too.

1. Introduction

The discovery of the phenomenon of perpendicular anisotropy has gained much interest in recent years. It has opened new possibilities for memory applications. Ferromagnetic thin films have been widely investigated for use as a data storage medium in magneto-optic or magneto-recording systems [1]. As the bandwidth of magneto-electronic devices approaches microwave frequencies, the role of damping in the switching response of magnetic thin films becomes ever more important. Details of the intrinsic mechanism underlying damping in ferromagnetic thin films remain unclear, despite extensive study [2–6].

The direction of the magnetization of thin ferromagnetic films depends on various anisotropic energy contributions like surface anisotropy fields which often favour an orientation perpendicular to the film, dipole interaction which favours an in-plane magnetization, and eventually anisotropy fields in the inner layers. In particular, with increasing temperature a reversible perpendicular to in-plane rotation as well as the reverse reorientation was observed. The magnetic anisotropy out of plane as well as in plane plays an important role in potential applications including half metallic magnetic tunnel junctions and magnetic perpendicular recording, as well as in fundamental investigations of magnetoresistance. The direction of magnetization relative to the body that supports it is determined mainly by two effects, shape anisotropy and magnetocrystalline anisotropy. The first arises from magnetostatic

effects and the second from spin–orbit coupling between the spins and the lattice of the material. The magnetostatic effects can be calculated from micromagnetic calculations, but the magnetocrystalline anisotropy must be computed from the electronic structure of the material.

Magnetic domains in ferromagnetic materials are generated in order to minimize the sum of energy terms, e.g., the magnetostatic, the exchange, the anisotropy, and the Zeeman energies. In bulk magnetic materials the magnetostatic coupling is normally small compared to the exchange interaction. In the array of magnetic elements the exchange interaction between the different elements is absent and therefore the magnetic properties of the array are governed by the magnetostatic coupling [7]. As has been shown both experimentally and theoretically, in patterned magnetic nanostructures the magnetostatic inter-element coupling can play an essential role and strongly affect their magnetic properties. Where the magnetic film has infinite lateral extension and is uniformly magnetized in the film plane, there is a demagnetizing field for a film. This magnetostatic field in a film with infinite lateral dimensions is not uniform close to the surface [45, 46]. So there is a non-zero magnetostatic energy in thin films and magnetostatic problems can be considered from discrete or semicontinuum models. If the film has a finite size, the surface charging has to be taken into account, which leads to a demagnetizing field. In the present paper the effects of magnetostatic interactions are neglected.

There is increasing activity in experimental [8–12] and theoretical [13–20] investigations to study the magnetic anisotropy in thin magnetic films. The main anisotropy contributions in thin films are the dipole interaction and the surface or interface lattice anisotropy due to the broken symmetry [21]. In addition, since most magnetic thin films on top or in between nonmagnetic material are distorted due to lattice mismatch, also the film interior layers may exhibit a strong volume anisotropy comparable to the surface anisotropy, which should not be neglected [22]. Magnetic anisotropies exhibit a strong temperature dependence mainly through the magnetization [23, 24]. Furthermore, these effective anisotropies are layer dependent, since for thin films the relative magnetization itself is layer dependent [25]. Usually, the magnetic anisotropy constants in thin films are found experimentally to be larger than that of the bulk materials [9, 10, 25–28]. Recently, Brune and co-workers [47] have introduced a method enabling the identification of the remarkably different contributions of surface and perimeter atoms to the magnetic anisotropy energy of two-dimensional nanostructures. They have shown for Co nanostructures on a Pt(111) surface that their uniaxial out-of-plane magnetization is entirely caused by edge atoms having 20 times more anisotropy energy than their bulk and surface counterparts. Identification of the role of perimeter and surface atoms opens up unprecedented opportunities for materials engineering. Another quite interesting series of results on magnetic surface and interface anisotropy with different interfaces came from Moessbauer experiments made by Shinjo and co-workers [48] and Walker and co-workers [49]. These authors observed quite numerous magnetic reconstructions close to the magnetic surface in different iron based and cobalt based cases.

Theoretically, it has been shown that the anisotropy coefficients $K_n(T)$, which are generally temperature dependent, can be calculated numerically at finite temperature within mean field theory, starting from a Hamiltonian with microscopic constant anisotropy parameters [14]. Strong anisotropies as compared to the exchange coupling and strongly different surface and volume anisotropies may induce a noncollinear thin film magnetization [15]. The thermal variation of the magnetization in ferromagnetic thin films is calculated by Pinettes and Lacroix [13] using the Holstein–Primakoff transformation which is valid for $T \leq T_C/3$. Jensen and Bennemann [16] obtained the temperature driven continuous and discontinuous reorientation of the magnetization in thin ferromagnetic films. Froeblich *et al* [17] presented a formal theory for the magnetization of thin ferromagnetic films on the basis of many-body Green’s function theory within a Heisenberg model with anisotropies. The

single-ion anisotropy terms are treated exactly by introducing higher-order Green's functions. The behaviour of ferromagnetic systems with single-ion anisotropies in more than one direction is investigated by Wang *et al* [18] with many-body Green's function theory. The effects of the exchange anisotropy and the single-ion anisotropy on the magnetic properties of thin ferromagnetic films are compared by Froebrich and Kuntz [19]. Schwieger *et al* [20] have considered the spin reorientation transition in a ferromagnetic Heisenberg monolayer with a second-order single-ion anisotropy as a function of temperature and external field. Using a Green-function decoupling method for spin 1/2 and the Heisenberg Hamiltonian taking into account the surface anisotropy coupling, Levy *et al* [29] have introduced the idea of 'hard' and 'soft' surfaces in ferromagnetic thin films according to their surface anisotropy, with a possible magnetic rearrangement close to the surface. A complementary study was reported by Levy [30] about ultra-thin films. First principles relativistic study of spin waves in thin magnetic films based on the adiabatic approach is presented in [31]. It is shown that the magnetocrystalline anisotropy energy contains contributions from both the on-site anisotropy surfaces and the off-site exchange coupling terms. The magnetic anisotropy energy and the interlayer exchange coupling of prototype trilayers are calculated using an *ab initio* approach based on the experimental lattice spacing [32].

In this paper we examine the single-ion anisotropy as a factor which contributes to the magnetic anisotropy and influences the static and dynamic properties of ferromagnetic thin films. To our knowledge the influence of the anisotropies on the damping has not been studied theoretically till now. We obtain some competition between the surface single-ion anisotropy and the surface exchange interaction.

2. Model and matrix Green's function

We consider a three-dimensional ferromagnetic system on a simple cubic (sc) lattice composed of N layers in the z -direction. The layers are numbered by $n = 1 \cdots N$, where the layers $n = 1$ and $n = N$ represent the two surfaces of the system. The bulk is established by the other layers. We start with the Hamiltonian of the Heisenberg model including a single-ion uniaxial anisotropy and an external magnetic field:

$$H_M = -\frac{1}{2} \sum_{l,\delta} J_{l,l+\delta} \mathbf{S}_l \mathbf{S}_{l+\delta} + \sum_i D_i (S_i^z)^2 - g\mu_B H_0 \sum_l S_l^z, \quad (1)$$

where the first term represents the isotropic exchange interactions and the second the single-ion anisotropic interactions. The exchange constants J and D are supposed to be positive and negative, respectively. The parameter J_{ij} is an exchange interaction between spins at nearest-neighbour sites i and j . To take into account the effects originated by the finite thickness of the system, we introduce two interaction parameters J_b and J_s . In the case of an interaction between spins, situated at the surface layer, the interaction strength is denoted by $J_{ij} = J_s$. Otherwise, the interaction in the bulk material is written as J_b , which is for simplicity assumed to be the same for the inter-layer coupling between the surface layer and the bulk as well as the intra-layer coupling between the different layers in the bulk. A similar notation is used for the single-ion anisotropy parameter $D_i = D_s$ for the surface and $D_i = D_b$ for the bulk. H_0 is a static magnetic field applied in the z direction.

In order to study the magnon excitations of the film we introduce the following retarded Green's function:

$$G_{ij}(t) = \langle\langle S_i^+(t); S_j^-(0) \rangle\rangle, \quad (2)$$

where S^+ and S^- are the spin operators. On introducing the two-dimensional Fourier transform $G_{n_i n_j}(\mathbf{k}_{\parallel}, E)$, one has the following form:

$$\langle\langle S_i^+; S_j^- \rangle\rangle_E = \frac{2\langle S^z \rangle}{N'} \sum_{\mathbf{k}_{\parallel}} \exp(i\mathbf{k}_{\parallel}(\mathbf{r}_i - \mathbf{r}_j)) G_{n_i n_j}(\mathbf{k}_{\parallel}, E), \quad (3)$$

where N' is the number of sites in any of the lattice planes, \mathbf{r}_i and n_i represent the position vectors of site i and the layer index, respectively, and $\mathbf{k}_{\parallel} = (k_x, k_y)$ is a two-dimensional wavevector parallel to the surface. The summation is taken over the Brillouin zone.

For the approximate calculation of the Green's function (2) we use a method proposed by Tserkovnikov [33], which is appropriate for spin problems. As a result the equation of motion for the Green's function (3) of the ferromagnetic thin film for $T \leq T_c$ has the following matrix form:

$$\mathbf{H}(E)\mathbf{G}(\mathbf{k}_{\parallel}, E) = \mathbf{R}, \quad (4)$$

where $\mathbf{H}(E)$ can be expressed as

$$\mathbf{H}(E) = \begin{pmatrix} E - v_1 + i\Gamma_1 & k_1 & 0 & 0 & 0 & 0 & \dots \\ k_2 & E - v_2 + i\Gamma_2 & k_2 & 0 & 0 & 0 & \dots \\ 0 & k_3 & E - v_3 + i\Gamma_3 & k_3 & 0 & 0 & \dots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \ddots \\ 0 & 0 & 0 & 0 & 0 & k_N & E - v_N + i\Gamma_N \end{pmatrix}$$

with

$$k_n = J\langle S_n^z \rangle, \quad n = 1, \dots, N,$$

$$v_n = 4J_n\langle S_n^z \rangle(1 - \gamma(\mathbf{k}_{\parallel})) + J_{n-1}\langle S_{n-1}^z \rangle + J_{n+1}\langle S_{n+1}^z \rangle + 2(J_{n-1} + 2D_{n-1})\langle S_{n-1}^z \rangle\langle S_n^z \rangle + 2(J_{n+1} + 2D_{n+1})\langle S_{n+1}^z \rangle^2 + 2D_n\langle S_n^z \rangle,$$

$$\Gamma_n = \frac{2\pi\langle S_n^z \rangle^2}{N'^2} \sum_{\mathbf{q}_{\parallel}\mathbf{p}_{\parallel}} [V_n^2(\mathbf{k}_{\parallel}, \mathbf{q}_{\parallel}, \mathbf{p}_{\parallel}) + D_n^2] [\bar{n}_{\mathbf{p}_{\parallel}}^n (1 + \bar{n}_{\mathbf{k}_{\parallel}-\mathbf{q}_{\parallel}}^n + \bar{n}_{\mathbf{p}_{\parallel}+\mathbf{q}_{\parallel}}^n) - \bar{n}_{\mathbf{k}_{\parallel}-\mathbf{q}_{\parallel}}^n \bar{n}_{\mathbf{p}_{\parallel}+\mathbf{q}_{\parallel}}^n] \delta(E_{\mathbf{p}_{\parallel}+\mathbf{q}_{\parallel}}^n + E_{\mathbf{k}_{\parallel}-\mathbf{q}_{\parallel}}^n - E_{\mathbf{p}_{\parallel}}^n - E_{\mathbf{k}_{\parallel}}^n),$$

$$V_n(\mathbf{k}_{\parallel}, \mathbf{q}_{\parallel}, \mathbf{p}_{\parallel}) = (J_{\mathbf{q}_{\parallel}} + J_{\mathbf{k}_{\parallel}-\mathbf{q}_{\parallel}-\mathbf{p}_{\parallel}}) - (J_{\mathbf{k}_{\parallel}-\mathbf{q}_{\parallel}} + J_{\mathbf{p}_{\parallel}+\mathbf{q}_{\parallel}}),$$

$$\gamma(\mathbf{k}_{\parallel}) = \frac{1}{2}(\cos(k_x a) + \cos(k_y a)).$$

$\langle S_n^z \rangle$ is the spin magnetization. $\bar{n}_{\mathbf{q}_{\parallel}} = \langle S_{\mathbf{q}_{\parallel}}^+ S_{\mathbf{q}_{\parallel}}^- \rangle$ is the spin correlation function which is calculated via the spectral theorem. $E(\mathbf{k}_{\parallel})$ is the spin-wave energy, which is calculated in the random phase approximation.

In order to obtain the solutions of the matrix equation (4), we define two-dimensional column matrices \mathbf{G}_n with the elements given by $(\mathbf{G}_n)_m = G_{mn}$ and $(\mathbf{R}_n)_m = 2\langle S_n^z \rangle \delta_{mn}$, so that equation (4) yields:

$$\mathbf{H}(E)\mathbf{G}_n = \mathbf{R}. \quad (5)$$

From equation (5), $G_{nn}(E)$ is obtained as

$$G_{nn}(E) = \frac{|H_{nn}(E)|}{|H(E)|}, \quad (6)$$

where $|H_{nn}(E)|$ is the determinant made by replacing the n th column of the determinant $|H(E)|$ by \mathbf{R}_n . The poles E_n of the Green's function $G_{nn}(E)$ can be obtained by solving $|H(E)| = 0$.

The thermal average of a spin in the n th layer for arbitrary magnitude of S is given by [34]

$$M_n = \langle S_n^z \rangle = \frac{1}{N'} \sum_{\mathbf{k}_{\parallel}} [(S + 0.5) \coth[(S + 0.5)\beta E_n] - 0.5 \coth(0.5\beta E_n)]. \quad (7)$$

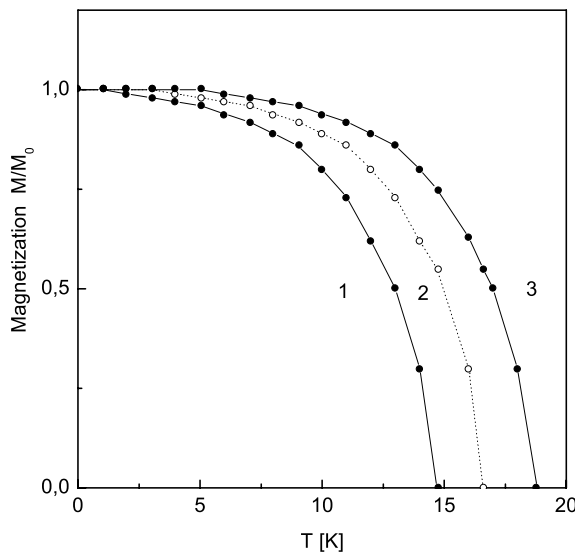


Figure 1. Temperature dependence of the magnetization M for a ferromagnetic thin film with $J_b = J_s = 0.4$ eV, $D_b = -0.1$ eV, $H = 0$, $N' = 2500$, $N = 7$ and different D_s -values: (1) $D_s = -0.05$, (2) -0.1 , (3) -0.3 eV.

3. Numerical results and discussion

In this section we shall present the numerical calculations of our theoretical results taking the following model parameter: $J_b = 0.4$ eV, $D_b = -0.1$ eV, $S = 3/2$. We have calculated the temperature dependence of the magnetization, the phase transition temperature, the spin-wave energies and the damping of the thin film for different values of the magnetic surface anisotropy constants. One has to solve self-consistently the N coupled equations (7) to obtain the layer magnetization. To characterize the complete ferromagnetic system both quantities, the magnetization and the spin-wave energy, are averaged over the N layers. The results for film thickness $N = 7$ and different surface single-ion anisotropies D_s are presented in figures 1 and 2. For the case of small surface anisotropies, when $D_s < D_b$ and has the value $D_s = -0.05$ eV (compare figures 1, 2—curve 1) the magnetization (respectively the spin-wave energy) is smaller than the case of $D_s = D_b$ (see figures 1, 2 curve 2). The magnetization decreases with increasing temperature to vanish at the critical temperature T_C of the thin film. The critical temperature decreases due to the smaller D_s value. The small anisotropy case is in qualitative agreement with experimental data on thin magnetic films, as in Ag/Fe(d)/W [34] and Ni(d)/Re [35].

In any case, the bulk anisotropy is negligible with respect to the dominating interface contribution [9]. Usually, the magnetic anisotropy constants in thin films are found to be larger than that of the bulk materials [9, 10, 25]. Epitaxial CoPt₃(111) films exhibit strong perpendicular magnetic anisotropy and are therefore potential materials for high-density magneto-optical recording [26]. Bochi *et al* [27, 28] have obtained a strong surface magnetoelastic anisotropy in epitaxial Cu/Ni/Cu (001) sandwiches and epitaxial fcc (111) Co/Cu superlattices, respectively. For the case of strong surface anisotropies, where $D_s = -0.3$ eV (figures 1, 2—curve 3), i.e. $D_s > D_b$, the magnetization (respectively the spin-wave energy) is larger than in the case $D_s = D_b$. The T_C of the film is enhanced due to the presence of larger D_s values. This is the opposite behaviour compared to the case of $D_s = -0.05$ eV, $D_s < D_b$. Sulitanu and Brinza [11] have considered Ni–W films and obtained a large perpendicular anisotropy constant associated with relatively high saturation magnetization, which should be used as alternative media for ultrahigh density perpendicular

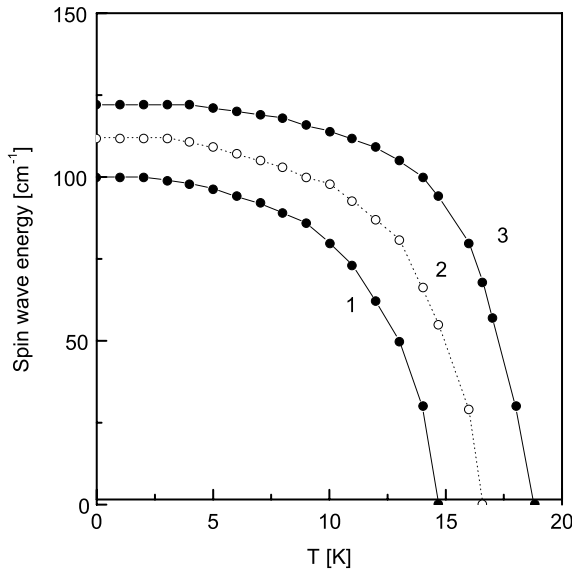


Figure 2. Temperature dependence of the spin-wave energy for a ferromagnetic thin film with $J_b = J_s = 0.4$ eV, $D_b = -0.1$ eV, $k = 0$, $H = 0$, $N = 7$ and different D_s -values: (1) $D_s = -0.05$, (2) -0.1 , (3) -0.3 eV.

magnetic recording. Our theoretical results for the strong anisotropy case are in qualitative agreement with the experimental data [9–11, 25–28].

From figures 1 and 2 we observe that the magnetization, the spin-wave energy and the critical temperature of the ferromagnetic phase transition are increased or decreased due to different surface anisotropy constants. But the changes of the physical properties cannot be described by a single parameter such as the surface magnetic anisotropy D_s . For example the concurrence between surface exchange interaction and surface magnetic anisotropy can be seen in figure 3. It is shown the influence of the surface exchange interaction constant J_s on the magnetization and the Curie temperature T_C . The results in figures 1 and 2 are obtained for $J_s = J_b$, i.e. the impact of the surface exchange interaction is negligible, which is not so realistic. But when the surface exchange interaction is smaller compared to the bulk value, $J_s \ll J_b$, then it would also contribute to the decrease of the Curie temperature T_C . If the influence of J_s is greater than that of D_s (figure 3 curve 1), then the magnetization, the spin-wave energy and T_C can be smaller than the bulk values although $D_s > D_b$ (compare figure 1, curve 3). With decreasing of J_s we obtain some coexistence or competition between the surface exchange interaction and the surface anisotropy effects, which is not considered till now. There exists for $D_s > D_b$ and $D_s = \text{constant}$ a ‘critical’ value of J_s below which the Curie temperature is smaller in comparison to the bulk value. Analogously we can obtain that in the case of $D_s < D_b$ the Curie temperature of the thin film can increase, for example when $J_s \gg J_b$ and $D_s = \text{constant}$ the Curie temperature and the magnetization are greater compared to the bulk (figure 3 curve 2; compare figure 1, curve 1). There exists for $D_s < D_b$ and $D_s = \text{constant}$ a ‘critical’ value of J_s above which the Curie temperature is greater in comparison to the bulk value.

The ferromagnetic temperature T_C of the thin magnetic film is found to be shifted with respect to the critical temperature of the bulk crystal. Moreover, the temperature shift appeared to be surface anisotropy dependent (figure 4). For $D_s < D_b$ we obtain a decrease of T_C with decreasing film thickness, whereas for $D_s > D_b$ an increase. We obtain a similar behaviour for the magnetization and the spin wave energies, too.

We have calculated the temperature dependence of the damping from $\Gamma = \frac{1}{N} \sum_n \Gamma_n$ for a simple cubic thin film for different values of the surface magnetic anisotropy constants.

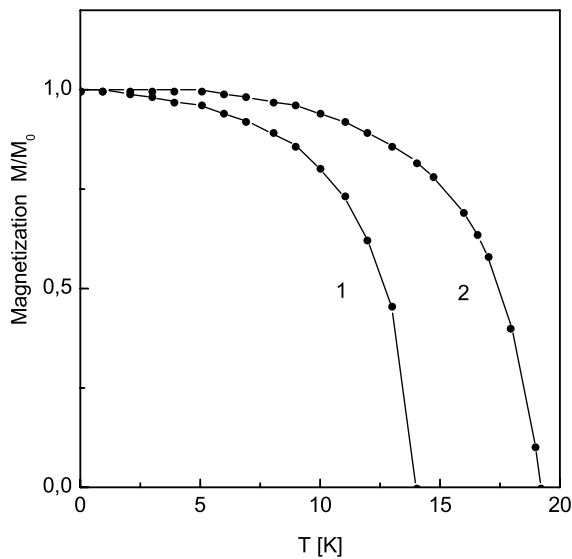


Figure 3. Temperature dependence of the magnetization M for a ferromagnetic thin film with $J_b = 0.4$ eV, $D_b = -0.1$ eV, $H = 0$, $N' = 2500$, $N = 7$ and different surface parameters: (1) $J_s = 0.1$, $D_s = -0.3$ eV (2) $J_s = 0.7$ eV, $D_s = -0.05$ eV.

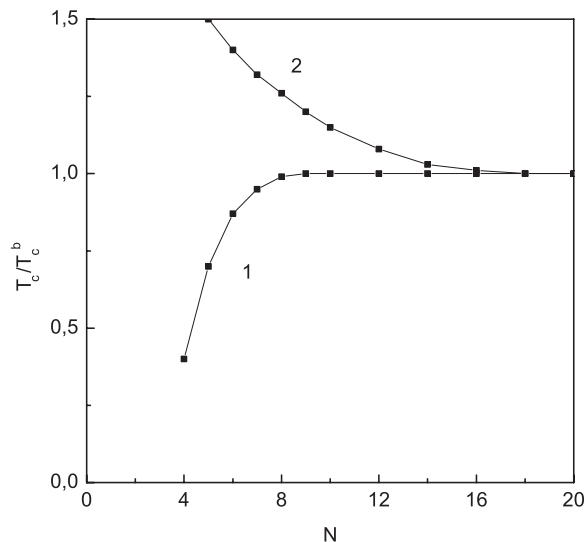


Figure 4. The dependence of the phase transition temperature T_C on the film thickness for $J_b = 0.4$ eV, $J_s = 0.2$ eV, $D_b = -0.1$ eV, $H = 0$ and different D_s -values: (1) $D_s = -0.05$; (2) -0.3 eV.

The results for film thickness $N = 7$ and different D_s -values are presented in figure 5. The damping increases with increasing temperature, $T \rightarrow T_C$, of the thin film. This could explain the observed experimental broadening of the resonance peaks in ferromagnetic thin films as the temperature approaches T_C [36]. It can be seen that the damping is always larger for both cases, small surface anisotropy $D_s < D_b$ (curve 2) and strong surface anisotropy $D_s > D_b$ (curve 3) in comparison to the case $D_s = D_b$ (curve 1). It increases due to the surface anisotropy effects. The damping, which is related to the linewidth in ferromagnetic resonance and Brillouin scattering experiments, increases with decreasing of the film thickness, which is in agreement with the experimental data [37, 38]. There are different mechanisms in thin films which contribute additively to the increasing of the damping such as surface effects [3], electron–phonon interactions [4], spin–phonon interactions [6], defects [5] and surface magnetic anisotropies which are considered in this paper.

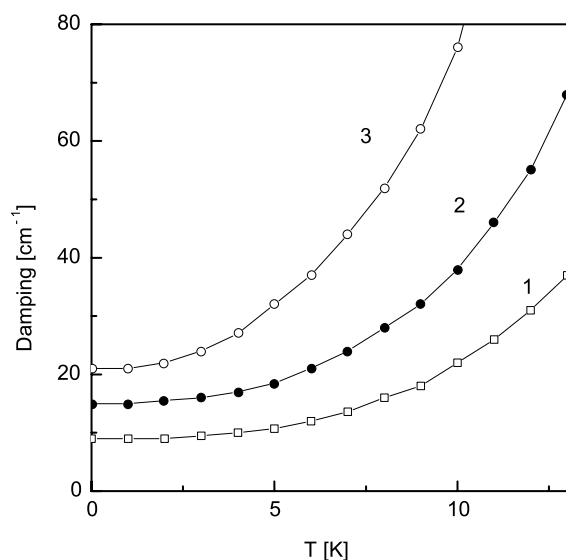


Figure 5. Temperature dependence of the damping Γ for a ferromagnetic thin film with $J_b = J_s = 0.4$ eV, $D_b = -0.1$ eV, $k = 0$, $H = 0$, $N = 7$ and different D_s -values: (1) $D_s = -0.1$; (2) -0.05 ; (3) -0.3 eV.

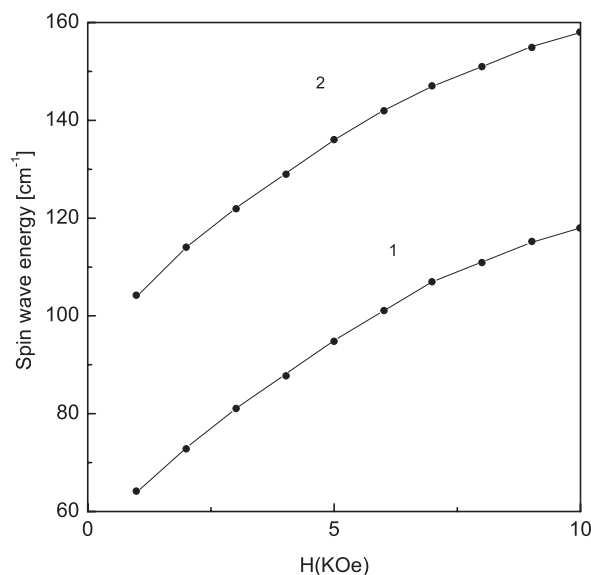


Figure 6. Spin-wave energies for ferromagnetic thin films as a function of the intensity of the external magnetic field H_0 for $J_b = 0.4$ eV, $J_s = 0.2$ eV, $D_b = -0.1$ eV, $D_s = -0.05$ eV and different film thicknesses: (1) $N = 7$; (2) 11.

Figure 6 shows the dependence of the spin-wave energy on the external magnetic field H_0 parallel to the magnetization for different film thicknesses. The spin-wave energy increases with increasing of H_0 and film thickness. This is in agreement with the experimental data of Tacchi *et al* [40]. If the external field is antiparallel to the magnetization direction, then the spin-wave energy decreases, in accordance with the experimental data of Acher *et al* [41].

The spin-wave damping Γ is numerically calculated in dependence on temperature, film thickness and magnetic field. At low temperatures, the damping is extremely small, smaller than the spin-wave energy, then with approaching of the critical temperature T_C it strongly increases [21]. Figure 7 demonstrates the film thickness dependence of the damping for different magnetic fields. For thinner films the damping is larger in comparison to thicker films. With increasing of the intensity of the external magnetic field the damping increases, too. The

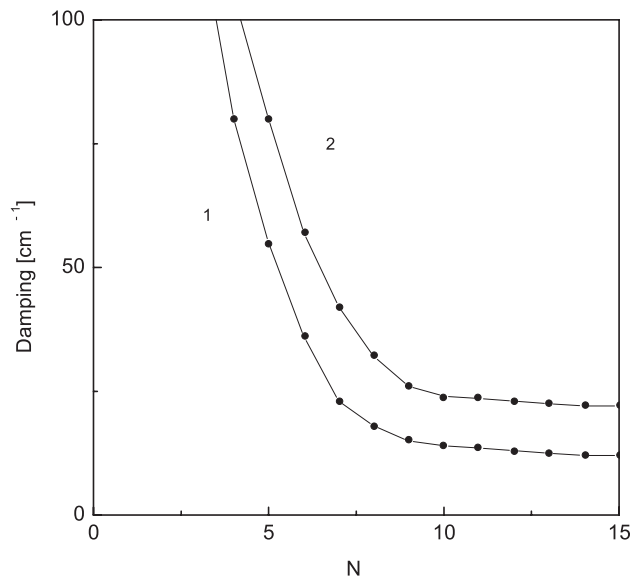


Figure 7. Film thickness dependence of the spin-wave damping Γ for $J_b = 0.4$ eV, $J_s = 0.2$ eV, $D_b = -0.1$ eV, $D_s = -0.05$ eV, $k = 0$ and for different external magnetic fields: (1) $H_0 = 5$ kOe, (2) 10 kOe.

obtained results are in good qualitative agreement with the experimental data of Azevedo *et al* [42], Rezende *et al* [43] and Fermin *et al* [44].

4. Conclusions

Using a Green's function technique the magnetization, the phase transition temperature, the spin-wave energy and the damping for ferromagnetic thin films are calculated and discussed for different temperatures, different surface magnetic anisotropy constants D_s in comparison to the bulk value D_b and different external magnetic fields. The magnetization and T_C decrease for $D_s < D_b$ and increase for $D_s > D_b$, respectively. The spin-wave energies are reduced in the first case and enhanced in the second case. The influence of the surface magnetic anisotropy constant on the transverse damping is obtained for the first time. It is shown that D_s can induce strong increasing of the damping in both cases of small or strong surface anisotropy. With increasing of the intensity of the external magnetic field the damping increases, too. The damping in thin films is greater compared to the bulk case due to different mechanisms which contribute additively to the damping, due to surface and substrate effects, electron-phonon interactions, spin-phonon interactions, defects, external magnetic fields and surface magnetic anisotropies which are considered in this paper.

It is shown that the changes of the physical properties cannot be described by a single parameter such as the surface magnetic anisotropy constant. We have seen that if we include a third effect, for example the influence of the surface exchange interaction, we can obtain the reverse result. The changes of the properties are the result of competition or coexistence between various mechanisms due to surfaces, film thickness, defects, spin-phonon interactions, substrates etc, and all must be taken into account in order to obtain correct results and if we want to explain the experimental data.

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