Influence of the spin–phonon interaction on the dynamical properties of ferromagnetic semiconducting thin films

2005 J. Phys.: Condens. Matter 17 6687

(http://iopscience.iop.org/0953-8984/17/42/009)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **17** (2005) 6687–6700 doi:10.1088/0953-8984/17/42/009

Influence of the spin–phonon interaction on the dynamical properties of ferromagnetic semiconducting thin films

J M Wesselinowa

Department of Physics, University of Sofia, Boulevard J Bouchier 5, 1164 Sofia, Bulgaria

Received 5 July 2005, in final form 30 August 2005
Published 7 October 2005
Online at stacks.iop.org/JPhysCM/17/6687

Abstract
The effects of spin–phonon interaction on the temperature dependence of the spin-wave and phonon spectrum in ferromagnetic semiconducting thin films are studied using a Green's function formalism beyond the random phase approximation. It is shown that due to the surface modes and the anharmonic spin–phonon interaction the spin-wave damping effects in thin ferromagnetic films are enhanced in comparison to the bulk. The phonon spectrum is discussed, too. Additional phonon damping and phonon frequency shift arise when the spin–phonon interaction is properly included.

1. Introduction

Ferromagnetic resonance (FMR) and Brillouin light-scattering (BLS) techniques have been used to investigate the spin-wave damping in ferromagnetic thin films [1]. The physical nature of the line width of ferromagnetic resonance absorption or BLS spectra is a long-standing subject and one of the most interesting problems in condensed matter. In some cases it is a difficult task to interpret experimental data with current theories involving elementary excitations and their microscopic interactions. Actually, the study of resonance line broadening is one of the main procedures for investigating the relaxation mechanisms and the laws governing the basic types of interactions in ferromagnetic systems. Indeed, FMR has been extensively used as a powerful tool for investigating the crystallinity and anisotropy properties of magnetic materials and, in particular, for studying the surface and interface phenomena associated with thin magnetic structures. The line widths measured in FMR and BLS spectra provide direct information on the spin-wave damping, or relaxation rate, in magnetic materials. There is current interest in the use of very thin films of ferromagnetic materials such as Fe as the basis for high frequency microwave devices. Many of the key properties in such devices depend on the strength of the damping in these systems. The relaxation (damping) of magnetization motion is a result of microscopic field fluctuations on spins by means of interaction with a thermal bath. These fluctuations appear due to either elementary processes, e.g., spin–spin, spin–electron, spin–phonon, or more complicated microscopic processes, such as slowly relaxing impurities, so that the line width is the combined effect of defects (or impurities) and damping. Each microscopic relaxation mechanism predicts its own temperature and frequency dependences of FMR and BLS line width. The magnetization damping can also depend on defect/impurity concentration and the sample size. It has been observed [1–5] that in general the FMR and BLS line width increases substantially as the film thickness decreases below certain values. On the other hand, Arias and Mills [6] made theoretical predictions for the extrinsic contribution to the FMR line width arising from the two-magnon scattering processes. Two-magnon scattering is a well known relaxation mechanism in bulk samples, which has also been shown to be present in thin films. Dobin and Victora [7] have shown that an important mechanism of ferromagnetic damping in thin films is the intrinsic four-magnon scattering process, which transfers energy from the initially excited uniform precession mode to $k = 0$ magnons.

Whereas the dynamical magnetic properties of thin magnetic films have been extensively theoretically investigated [8, 9], it is not so with ferromagnetic semiconducting thin films. Gopalan and Cottam [10] have used the s–d interaction model to study the bulk and surface magnetic excitations of a semi-infinite ferromagnetic semiconductors for low temperatures. Mills [11] has studied the ferromagnetic resonance relaxation in ultrathin metal films, and specially the role of the conduction electrons. The temperature dependence of the layer magnetization and the thickness dependence of the Curie temperature of thin ferromagnetic semiconducting films in the ferromagnetic phase are investigated within the s–d model and a Green's function formalism by Wesselinowa *et al* [12]. The dynamical properties of thin ferromagnetic semiconducting films are obtained by Wesselinowa [13]. It is shown that the frequencies of the films are smaller, whereas the damping effects are larger compared to the bulk.

During the last two decades an impressive development of experimental techniques has been achieved to even pin down the fundamental interactions in solids, such as the electron–electron [14], electron–phonon [15, 16] and electron–magnon interactions [17]. The effect of electron–phonon interaction on the temperature dependence of the electronic spectrum and damping in thin ferromagnetic semiconducting films is studied theoretically by Wesselinowa [18]. The phonon spectrum is discussed, too. Additional phonon damping and phonon frequency shift arise when the electron–phonon interaction is properly included. Phonon–magnon scattering is considered as a major relaxation mechanism of excitations in a ferromagnet [19, 20]. McMichel and Kunz [21] have calculated the ferromagnetic resonance damping rates due to coupling between the magnetization and lattice vibrations through inhomogeneities. Booth *et al* [22] have established the important relationship between the electronic, spin and phonon systems.

The temperature dependence of lifetime broadening of the Gd(0001) surface state is studied using scanning tunnelling spectroscopy [23]. The coherent surface phonon at a GaAs surface has been investigated by time-resolved second-harmonic generation [24, 25]. The frequency of the surface component shows red shifts as the pumping power increases. The shifts are indicative of a marked electron–phonon interaction or anharmonicity of the surface phonon modes. The phonon density of states (DOS) in thin films of Fe was measured by inelastic nuclear resonant scattering of synchrotron radiation [26]. The thin-film DOS exhibits significant deviations from the DOS of bulk Fe, which the authors attribute to phonon lifetime broadening in the confined geometry. Generally, the anharmonicity of surface phonon modes is considered to be greater than that of bulk phonon modes. Baddorf and Plummer [27] have revealed that the anharmonicity for the motion normal to the surface on a Cu(110) surface is four to five times greater than that in bulk copper. Theoretical studies of the surface phonon linewidth of Ag, Cu and Al are presented by Rahman *et al* [28].

The purpose of the present paper is to investigate the spin–phonon interaction in ferromagnetic semiconducting thin films on the basis of the s–d interaction model. For the calculation we use the method of the retarded Green's functions and especially the method of Tserkovnikov [29], which is appropriate for spin problems. In this context it is worthwhile to mention that this method can be applied for various materials with complex magnetic interactions—metals, rare-earth metals, magnetic semiconductors, metal–insulator systems, magnetoelectric systems etc. We can calculate different magnetic, electronic, optical and transport properties, i.e. different static and dynamic properties (for example damping and relaxation times). We need a model with a quantum mechanical Hamiltonian which contains different interactions—spin–spin, s–d(f), spin–phonon, dipole–dipole, spin–orbit etc. The method can be applied to different dimensional systems, such as bulk materials, thin films, nanoparticles and chains.

2. The model

Let us first introduce the model. We consider a three-dimensional ferromagnetic semiconducting system on a simple cubic (sc) lattice composed of N layers in the z-direction. The layers are numbered by $n = 1 \ldots N$, where the layers $n = 1$ and N represent the two surfaces of the system. The bulk is established by the other layers. To take into account specific surface effects we start with the Hamiltonian of the s–d model including both bulk and surface properties:

$$H = H_{\mathrm{M}} + H_{\mathrm{E}} + H_{\mathrm{ME}} + H_{\mathrm{P}} + H_{\mathrm{SP}}. \tag{1}$$

H_{M} is the Heisenberg Hamiltonian for the ferromagnetically ordered d electrons:

$$H_{\mathrm{M}} = -\frac{1}{2}\sum_{l,\delta} J_{l,l+\delta}\mathbf{S}_l\mathbf{S}_{l+\delta} + \sum_i D_i (S_i^z)^2, \tag{2}$$

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 single-ion anisotropy parameter is typically smaller by some orders of magnitude than the Heisenberg exchange interaction, $|D_i| \ll J_{ij}$.

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 and J_{s}. In the case of an interaction between spins, situated at the surface layer, the interaction strength is denoted by $J_{ij} = J_{\mathrm{s}}$. Otherwise, the interaction in the bulk material is written as J, 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 all parameters in equations (2)–(7).

H_{E} represents the usual Hamiltonian of the conduction band electrons,

$$H_{\mathrm{E}} = \sum_{l,\delta,\sigma} t_{l,l+\delta} c_{l\sigma}^{\dagger} c_{l+\delta,\sigma}, \tag{3}$$

where $t_{l,l+\delta}$ is the hopping integral.

H_{ME} couples the two subsystems (2) and (3) by an intra-atomic exchange interaction I_l,

$$H_{\mathrm{ME}} = -\sum_l I_l \mathbf{S}_l \mathbf{s}_l. \tag{4}$$

The spin operators $\mathbf{s}_l$ of the conduction electrons at site l can be expressed as $s_l^+ = c_{l+}^{\dagger} c_{l-}$, $s_l^z = (c_{l+}^{\dagger} c_{l+} - c_{l-}^{\dagger} c_{l-})/2$, where $c_{l\sigma}^{\dagger}$ and $c_{l\sigma}$ are Fermi creation and annihilation operators at site l, respectively; $\sigma = \pm 1$ corresponds to spin-up and down states.

H_P contains the lattice vibrations including third- and fourth-order anharmonic phonon–phonon interactions:

$$H_P = \frac{1}{2!}\sum_{\mathbf{q}}(P_{\mathbf{q}}P_{-\mathbf{q}} + \omega_{\mathbf{q}}^2 Q_{\mathbf{q}}Q_{-\mathbf{q}}) + \frac{1}{3!}\sum_{\mathbf{q}\mathbf{q}_1} B(\mathbf{q},\mathbf{q}_1)Q_{\mathbf{q}}Q_{-\mathbf{q}_1}Q_{\mathbf{q}_1-\mathbf{q}}$$
$$+ \frac{1}{4!}\sum_{\mathbf{q},\mathbf{q}_1,\mathbf{q}_2} A(\mathbf{q},\mathbf{q}_1,\mathbf{q}_2)Q_{\mathbf{q}_1}Q_{\mathbf{q}_2}Q_{-\mathbf{q}-\mathbf{q}_2}Q_{-\mathbf{q}_1+\mathbf{q}}, \tag{5}$$

where $Q_{\mathbf{q}\lambda}$, $P_{\mathbf{q}\lambda}$ and $\omega_{\mathbf{q}}$ are the normal coordinate, momentum and frequency, respectively, of the lattice mode with a wavevector $\mathbf{q}$. The vibrational normal coordinate $Q_{\mathbf{q}}$ and the momentum $P_{\mathbf{q}}$ can be expressed in terms of phonon creation and annihilation operators: $Q_{\mathbf{q}\lambda} = (2\omega_{\mathbf{q}\lambda})^{-1/2}(a_{\mathbf{q}\lambda} + a^{\dagger}_{-\mathbf{q}\lambda})$, $P_q = \mathrm{i}(\omega_{\mathbf{q}\lambda}/2)^{1/2}(a^{\dagger}_{\mathbf{q}\lambda} - a_{-\mathbf{q}\lambda})$.

The final term H_{SP} represents interactions between the spin and the phonon systems including anharmonic terms:

$$H_{SP} = -\tfrac{1}{2}\sum_{\mathbf{q}\mathbf{p}} F(\mathbf{q},\mathbf{p})Q_{\mathbf{p}-\mathbf{q}}(S_q^z S_{-p}^z + S_q^- S_p^+)$$
$$- \tfrac{1}{4}\sum_{\mathbf{q}\mathbf{p}\nu} R(\mathbf{q},\mathbf{p},\nu)Q_{\nu}Q_{\mathbf{p}-\mathbf{q}-\nu}(S_q^z S_{-p}^z + S_q^- S_p^+) + \text{h.c.} \tag{6}$$

3. The magnon Green's function

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

$$g_{ij}(t) = \langle\langle S_i^+(t); S_j^-(0)\rangle\rangle, \tag{7}$$

where S^+ and S^- are the spin-$\frac{1}{2}$ operators. On introducing the two-dimensional Fourier transform $g_{n_in_j}(\mathbf{k}_\|, \omega)$, one has the following form:

$$\langle\langle S_i^+; S_j^-\rangle\rangle_\omega = \frac{\sigma}{N'}\sum_{\mathbf{k}_\|}\exp(\mathrm{i}\mathbf{k}_\|(\mathbf{r}_i - \mathbf{r}_j))g_{n_in_j}(\mathbf{k}_\|, \omega), \tag{8}$$

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 $\mathbf{k}_\| = (k_x, k_y)$ 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 (8) we use a method proposed by Tserkovnikov [29], which is appropriate for spin problems. As a result the equation of motion for the Green's function (8) of the ferromagnetic semiconducting film for $T \leqslant T_C$ has the following matrix form:

$$\mathbf{L}(\mathbf{E})\mathbf{g}(\mathbf{k}_\|, E) = \mathbf{R}, \tag{9}$$

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

$$\mathbf{L}(\mathbf{E}) = \begin{pmatrix} E - L_1 + \mathrm{i}\Gamma_1 & k_1 & 0 & 0 & 0 & 0 & \dots \\ k_2 & E - L_2 + \mathrm{i}\Gamma_2 & k_2 & 0 & 0 & 0 & \dots \\ 0 & k_3 & E - L_3 + \mathrm{i}\Gamma_3 & k_3 & 0 & 0 & \dots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \ddots \\ 0 & 0 & 0 & 0 & 0 & k_N & E - L_N + \mathrm{i}\Gamma_N \end{pmatrix}$$

with

$$k = J\langle S_n^z\rangle, \qquad n = 1, \dots, N,$$

$$L_n = I_n\langle s^z\rangle_n + \frac{I_n^2\langle S_n^z\rangle\langle s_n^z\rangle}{\omega - I_n\langle S_n^z\rangle} + 4J_n\langle S_n^z\rangle(1 - \gamma(\mathbf{q}_\|)) + J_{n-1}\langle S_{n-1}^z\rangle + J_{n+1}\langle S_{n+1}^z\rangle,$$

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

$\langle S_n^z \rangle$ and $\langle s_n^z \rangle$ are the localized-spin and conduction electron magnetization, respectively [12, 22]. Calculations yield the following expression for the transverse spin-wave damping Γ_n^{s}:

$$\Gamma_n^{\mathrm{s}} = \Gamma_n^{\mathrm{ss}} + \Gamma_n^{\mathrm{sd}} + \Gamma_n^{\mathrm{sp}}. \tag{10}$$

To the transverse spin-wave damping Γ_n^{s} contribute the dampings Γ_n^{ss}, Γ_n^{sd} and Γ_n^{sp}, due to the spin–spin, s–d and spin–phonon interactions, respectively. The terms are given in appendix A.

The spin, electron and phonon correlation functions $\bar{n}_{\mathbf{q}_\parallel} = \langle S^+_{\mathbf{q}_\parallel} S^-_{\mathbf{q}_\parallel} \rangle$, $\bar{m}_{\mathbf{q}_\parallel \sigma} = \langle c^\dagger_{\mathbf{q}_\parallel \sigma} c_{\mathbf{q}_\parallel \sigma} \rangle$ and $\bar{N}_{\mathbf{q}_\parallel} = \langle a^\dagger_{\mathbf{q}_\parallel} a_{\mathbf{q}_\parallel} \rangle$ which appear in the damping terms are obtained via the spectral theorem. $E(\mathbf{k}_\parallel)$, $\epsilon(\mathbf{k}_\parallel)$ and $\bar{\omega}(\mathbf{k}_\parallel)$ are the renormalized spin-wave, electronic and phonon energies, respectively. For the calculation of the conduction band magnetization, the electronic energy and the electron correlation function it is necessary to define a one-electron Green's function by $G_\sigma(k) = \langle\langle c^\dagger_{k\sigma} c_{k\sigma} \rangle\rangle$ [18].

In order to obtain the solutions of the matrix equation (9), we define two-dimensional column matrices $\mathbf{g}_m$ 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 (9) yields

$$\mathbf{L}(\mathbf{E})\mathbf{g}_n = \mathbf{R}. \tag{11}$$

From equation (11), $g_{nn}(E)$ is obtained as

$$g_{nn}(E) = \frac{|L_{nn}(E)|}{|L(E)|}, \tag{12}$$

where $|L_{nn}(E)|$ is the determinant made by replacing the nth column of the determinant $|L(E)|$ by I. The poles E_n of the Green's function $g_{nn}(E)$ can be obtained by solving $|L(E)| = 0$.

4. The phonon Green's function

In order to obtain the phonon spectrum we have to define the phonon Green's function:

$$G_{ij}(t) = \langle\langle a_i(t); a_j^\dagger(0) \rangle\rangle. \tag{13}$$

Analogous to the previous section, after a two-dimensional Fourier transformation we get for the matrix Green's function the following expression:

$$\mathbf{H}(\omega)\mathbf{G}(\mathbf{k}_\parallel, \omega) = \mathbf{I}, \tag{14}$$

where

$$\mathbf{H}(\omega) = \begin{pmatrix} \omega - V_1 + \mathrm{i}\gamma_1 & k_1 & 0 & 0 & 0 & 0 & \cdots \\ k_2 & \omega - V_2 + \mathrm{i}\gamma_2 & k_2 & 0 & 0 & 0 & \cdots \\ 0 & k_3 & \omega - V_3 + \mathrm{i}\gamma_3 & k_3 & 0 & 0 & \cdots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \ddots \\ 0 & 0 & 0 & 0 & 0 & k_N & \omega - V_N + \mathrm{i}\gamma_N \end{pmatrix}$$

with

$$k_n = R(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \mathbf{k}_\parallel)\langle S^z \rangle_n^2 - B(\mathbf{k}_\parallel, \mathbf{k}_\parallel, 0)\langle Q_{\mathbf{k}_\parallel} \rangle \delta_{\mathbf{k}_\parallel 0},$$

$$\begin{aligned} V_n = \omega_{\mathbf{k}_\parallel} - \frac{1}{N'} \sum_{\mathbf{q}_\parallel} \Big[& R_n(\mathbf{k}_\parallel, \mathbf{q}_\parallel, \mathbf{q}_\parallel)\langle S^z \rangle_n^2 + R_{n-1}(\mathbf{k}_\parallel, \mathbf{q}_\parallel, \mathbf{q}_\parallel)\langle S^z \rangle_{n-1}^2 \\ & + R_{n+1}(\mathbf{k}_\parallel, \mathbf{q}_\parallel, \mathbf{q}_\parallel)\langle S^z \rangle_{n+1}^2 \Big] + B_n(\mathbf{k}_\parallel, -\mathbf{k}_\parallel, 0)\langle Q_0 \rangle_n \\ & + B_{n-1}(\mathbf{k}_\parallel, -\mathbf{k}_\parallel, 0)\langle Q_0 \rangle_{n-1} + B_{n+1}(\mathbf{k}_\parallel, -\mathbf{k}_\parallel, 0)\langle Q_0 \rangle_{n+1} \end{aligned}$$

$$+\frac{1}{2N'}\Big[(A_n(\mathbf{k}_\parallel,-\mathbf{k}_\parallel,\mathbf{q}_\parallel,-\mathbf{q}_\parallel)+A_n(\mathbf{q}_\parallel,-\mathbf{k}_\parallel,-\mathbf{q}_\parallel,\mathbf{k}_\parallel))(2\bar{N}_n(\mathbf{q}_\parallel)+1)$$
$$+(A_{n-1}(\mathbf{k}_\parallel,-\mathbf{k}_\parallel,\mathbf{q}_\parallel,-\mathbf{q}_\parallel)+A_{n-1}(\mathbf{q}_\parallel,-\mathbf{k}_\parallel,-\mathbf{q}_\parallel,\mathbf{k}_\parallel))(2\bar{N}_{n-1}(\mathbf{q}_\parallel)+1)$$
$$+(A_{n+1}(\mathbf{k}_\parallel,-\mathbf{k}_\parallel,\mathbf{q}_\parallel,-\mathbf{q}_\parallel)+A_{n+1}(\mathbf{q}_\parallel,-\mathbf{k}_\parallel,-\mathbf{q}_\parallel,\mathbf{k}_\parallel))(2\bar{N}_{n+1}(\mathbf{q}_\parallel)+1)\Big],$$

$$\langle Q_{\mathbf{k}_\parallel}\rangle=\frac{F_{\mathbf{k}_\parallel,\mathbf{k}_\parallel}\langle S^z\rangle^2-\frac{1}{N'}\sum_{\mathbf{q}_\parallel}B_{\mathbf{k}_\parallel,\mathbf{q}_\parallel}(2\bar{N}_{\mathbf{q}_\parallel}+1)}{\omega_{\mathbf{k}_\parallel}-R_{\mathbf{k}_\parallel,\mathbf{k}_\parallel,\mathbf{k}_\parallel}\langle S^z\rangle^2+\frac{1}{N'}\sum_{\mathbf{q}_\parallel}A_{\mathbf{k}_\parallel,\mathbf{q}_\parallel}(2\bar{N}_{\mathbf{q}_\parallel}+1)}.$$

We obtain the following expression for the phonon damping taking into account the spin–phonon interaction:

$$\gamma_n^{\mathrm{ph}}=\gamma_n^{\mathrm{sp}}+\gamma_n^{\mathrm{ph-ph}}. \tag{15}$$

γ_n^{sp} is the damping part which comes from the spin–phonon interaction and $\gamma_n^{\mathrm{ph-ph}}$ is the phonon damping due to the phonon–phonon anharmonic interaction. The damping terms are given in appendix B.

We obtain the solutions of the matrix equation (14) analogous to the previous section.

5. Numerical results and discussion

We turn in this paper to the influence of the spin–phonon interaction, temperature and film thickness on the dynamical behaviour of thin films. Thin films are of particular interest because their critical properties are more susceptible to surface parameters than thicker films. The temperature dependence of the renormalized spin-wave energy is calculated numerically for a simple cubic (sc) thin ferromagnetic semiconducting film with parameters for $CdCr_2Se_4$ [30]: $J_s=0.2\,J$, $D_s=D=0.01$ eV, $I_s=0.2I$, $t_s=0.05t$, $F_s=2F$, $R_s=2R$, $B_s=2B$, $A_s=2A$, $J=0.1$ eV, $I=0.5$ eV, $t=0.1$ eV, $B=-2.54\ \mathrm{cm}^{-1}$, $F=23\ \mathrm{cm}^{-1}$, $R=-18\ \mathrm{cm}^{-1}$, $A=6.61\ \mathrm{cm}^{-1}$, $S=3/2$, $W=0.1$ eV, $\mathbf{k}=0$, $T_C=130$ K. At a solid surface, the crystal symmetry is broken, and the anharmonicity is expected to be a factor of two to three greater than in the bulk [31, 32]. Therefore we have chosen greater surface anharmonic constants compared to the bulk. It will be shown that the enhanced surface anharmonicity leads to a decrease in energy and increase in width of a surface phonon.

5.1. The spin-wave spectrum

The spin-wave energy and the damping were determined taking into account spin–phonon interactions from equation (9), by solving $|L(E)|=0$ (12). The temperature dependence of the spin-wave frequencies is plotted in figure 1 for a simple cubic ferromagnetic semiconducting film for $\mathbf{k}_\parallel=0$ and different thicknesses of the film ($N=8$ and 30 layers). It is found that there are several differences between the thin films and the bulk behaviour and between the quantities with and without spin–phonon interaction. With decreasing of the film thickness the frequency decreases, too. Furthermore the spin–phonon interaction decreases the spin-wave energies. We obtain that for $N<30$ layers is valid:

$$\epsilon_{\mathrm{TF}}<\epsilon_{\mathrm{B}}. \tag{16}$$

The spin-wave energy of the film with $N>30$ layers coincides with that for the bulk.

Now we will study the different contributions to the spin-wave damping. Firstly we consider the zero-temperature limit $T=0$. At $T=0$ the expression of the spin-wave damping of the ferromagnetic semiconducting thin film simplifies to

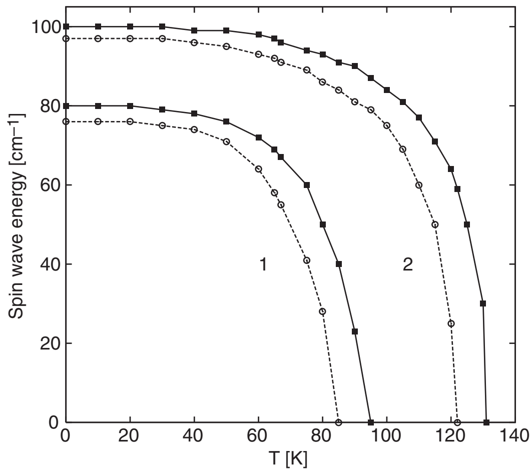

Figure 1. Temperature dependence of the spin-wave energy ϵ for an sc ferromagnetic semiconducting thin film for $J_s = 0.2J_b$, $I_s = I_b$, $B_s = 2B_b$, $A_s = 2A_b$, $F_s = 0.2F_b$, $R_s = 2R_b$, and different film thicknesses: (1) $N = 8$, (2) 30 layers; dashed line—with, full line—without spin–phonon interaction.

$$
\begin{aligned}
\Gamma_n^{s}(T=0) = {} & \frac{\pi}{4N'} \sum_{\mathbf{q}_\parallel \mathbf{q}_\parallel \mathbf{q}_\parallel} F_n^2(\mathbf{k}_\parallel, \mathbf{q}_\parallel)\delta\Big(E^n_{\mathbf{q}_\parallel} - \omega^n_{\mathbf{q}_\parallel - \mathbf{k}_\parallel} - E^n_{\mathbf{k}_\parallel}\Big) \\
& + \frac{\pi}{N'} \sum_{\mathbf{q}_\parallel \mathbf{p}_\parallel \mathbf{r}_\parallel} \Big[R_n^2(\mathbf{q}_\parallel, \mathbf{p}_\parallel, \mathbf{r}_\parallel) + R_n(\mathbf{q}_\parallel, \mathbf{p}_\parallel, \mathbf{r}_\parallel) R_n(\mathbf{p}_\parallel - \mathbf{q}_\parallel - \mathbf{r}_\parallel, \mathbf{p}_\parallel, \mathbf{q}_\parallel)\Big] \\
& * \delta(E^n_{\mathbf{k}_\parallel + \mathbf{q}_\parallel - \mathbf{p}_\parallel} + \bar{\omega}^n{}_{\mathbf{r}_\parallel} + \bar{\omega}^n_{\mathbf{p}_\parallel - \mathbf{q}_\parallel - \mathbf{r}_\parallel} + E^n_{\mathbf{k}_\parallel}).
\end{aligned} \tag{17}
$$

We can see that at $T = 0$ and low temperatures the spin waves are damped due to the spin–phonon interaction. At temperatures close to T_C and for $T \geqslant T_C$, $\Gamma^{ss} = 0$; i.e. the spin-wave damping in the high-temperature region is due to the s–d and s–p interaction.

Following the spin–phonon interaction plays an important role for low and high temperatures. At low temperatures γ^{sp} is very small. With increasing temperature, the damping γ^{sp} increases, and the contribution of the anharmonic terms increases, too. The term proportional to the spin–phonon constant F is nearly temperature independent. Hence the anharmonic terms give the main contribution to the temperature dependence of the spin-wave damping.

The temperature dependence of the spin-wave damping for a thin ferromagnetic semiconducting film with $N = 7$ is presented in figure 2. At low temperatures the damping is very small, the spin-wave energy is greater than the damping term. Approaching T_C, Γ^s increases strongly. The damping parts, due to the s–d and s–p interactions, predominate over this, which is due to the spin–spin interaction. Therefore we have

$$\Gamma^{ss} \ll \Gamma^{sp} \ll \Gamma^{s-d}. \tag{18}$$

The spin-wave damping as a function of temperature is plotted in figure 3 with and without spin–phonon interaction for various film thicknesses ($N = 8$ and 30 layers). Thinner films have larger damping. For $N < 30$ layers we have

$$\gamma_{TF} > \gamma_B, \tag{19}$$

i.e. γ is larger for thin films than that for the bulk. This is in agreement with the experimental data [1–5]. The spin–phonon interaction enhances the damping and contributes to the

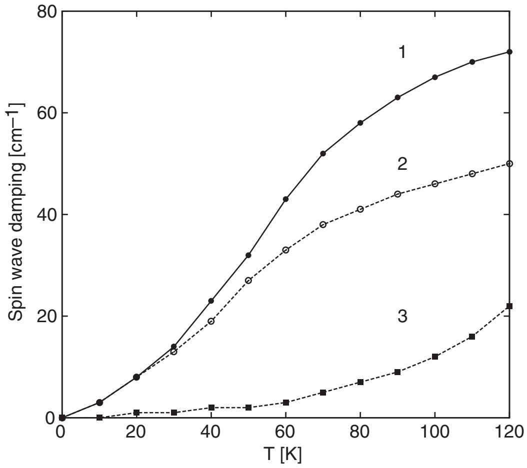

Figure 2. Temperature dependence of the spin-wave damping for $F = 23\ \mathrm{cm}^{-1}$, $R = -18\ \mathrm{cm}^{-1}$ and $\omega_0 = 100\ \mathrm{cm}^{-1}$: curve 1, total spin-wave damping Γ^{s}; curve 2, spin-wave damping Γ^{sd} from the s–d interaction; curve 3, the spin-wave damping Γ^{sp} due to the spin–phonon interaction.

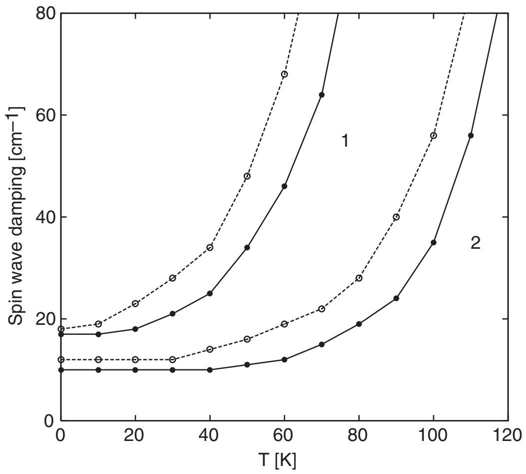

Figure 3. Temperature dependence of the spin-wave damping Γ_s for the same parameters as in figure 1, and different film thicknesses: (1) $N = 8$, (2) 30 layers; dashed line—with, full line—without spin–phonon interaction.

experimentally obtained line broadening. It must be taken into account in order to obtain correct results.

It is worth-while to mention that in our calculations we have not taken into account the influence of the surface single-ion anisotropic interaction constant; the exchange constants from surface and bulk are equal. But this is not always so. It is known that the surface anisotropies are important for thin films. This will be considered in a next paper. If we take $D_s \neq D_b$ then in the transverse spin-wave damping there appears an additional term due to the surface anisotropy, which increases the damping and contributes to the broadening of the line widths.

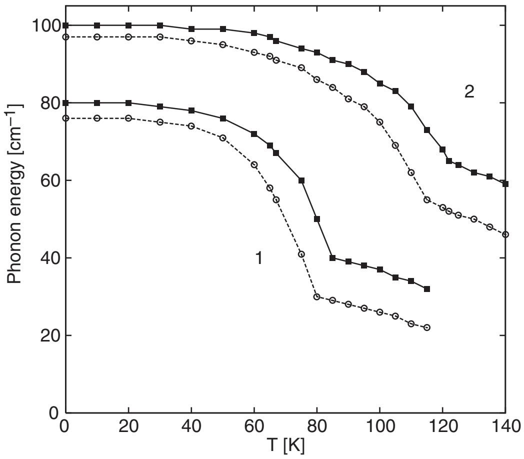

Figure 4. Temperature dependence of the phonon A mode with $\omega_0 = 100$ cm^{-1} for an sc ferroelectric film for the same parameters as in figure 1, and different film thicknesses: (1) $N = 8$, (2) 30 layers; dashed line—with, full line—without spin–phonon interaction.

5.2. The phonon spectrum

Now we will study the effects of the spin–phonon and phonon–phonon interactions on the phonon frequency and damping in ferromagnetic semiconducting thin films. The phonon energy and the damping were calculated numerically using the same parameters as for the spin-wave spectrum. Some interesting features are observed from the obtained results. The surface phonon energy $\bar{\omega}_s$ is much smaller than the phonon energy of the inner layer $\bar{\omega}_{N/2}$; the surface phonon decreases four times more than observed in the bulk. This is due to the lower coordination number of the surface phonons and to the spin–phonon interaction. With increasing of the spin–phonon interaction constant the surface phonon energy decreases. The surface damping Γ_s^{ph} is much larger compared to the damping of the inner layer $\Gamma_{N/2}^{ph}$. The big difference between the surface spectrum and the spectrum of the inner layer can be explained as the result of surface modes, which are damped quickly on going into the bulk due to the confined geometry, and due to the spin–phonon interaction.

The optical phonon energy $\bar{\omega}_k$ is renormalized owing to the anharmonic phonon–phonon and spin–phonon interactions. If they are not taken into account, then $\bar{\omega}_k$ is identical with the energy ω_k of the uncoupled optical phonon. It will be independent of temperature. We have studied the temperature dependence of the renormalized phonon frequencies using the same model parameters as in figures 1 and 2. The phonon mode displays a non-linear dependence on temperature when T approaches T_C. Since it is a lattice mode this behaviour can be described to strong anharmonic effects. If we take into account only the third-order interaction terms, i.e. $A = 0$, $B = 0$, then we obtain a linear temperature dependence close to T_C. It is evident that there is a strong anharmonicity affecting the phonon modes near the transition point from the ferroelectric to the paraelectric phase. The calculations demonstrate that we must not neglect the effects of the spin ordering, and the Hamiltonian which describes the system must include terms taking into account not only the anharmonic phonon–phonon interaction but also the anharmonic spin–phonon interaction. The temperature dependence of the renormalized phonon mode $\omega_0 = 100$ cm^{-1} (D mode) for $CdCr_2Se_4$ is plotted in figure 4 for different thicknesses of the film ($N = 8$ and 30 layers) with and without spin–phonon interaction. It can be seen that the spin–phonon interaction reduces the phonon energy and must be taken

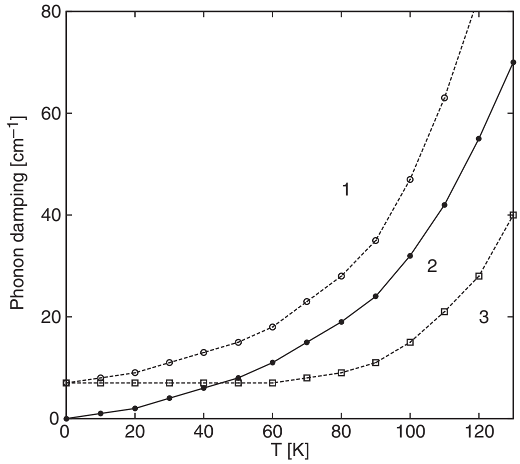

Figure 5. Temperature dependence of the phonon damping with $\omega_0 = 100$ cm^{-1} for the same parameters as in figure 4: total phonon damping γ^{ph}; curve 2, phonon damping γ^{ph-ph} including the anharmonic phonon–phonon interaction; curve 3, phonon damping γ^{sp} due to the spin–phonon interaction.

into account if we want to obtain correct results and to explain the experimental data. With increasing film thickness the frequency increases, too. For $N < 30$ layers we obtain that

$$\bar{\omega}_{TF} < \bar{\omega}_B, \tag{20}$$

i.e. the phonon frequency of the thin film shifts to lower energy due to the existence of a surface mode and due to spin–phonon coupling. The phonon energy of the film with $N > 30$ layers coincides with that for the bulk.

Analogous to section 5.1 we want to discuss the phonon damping on temperature, anharmonicity and film thickness. The temperature dependence of Γ^{ph} for $N = 8$ layers obtained using the same model parameters as in figure 4 is shown in figure 5. Firstly we consider the zero-temperature limit $T = 0$. At $T = 0$, where the part of the phonon damping due to the phonon–phonon interaction γ^{ph-ph} vanishes, we obtain

$$\begin{aligned}\gamma_n^{ph}(T=0) = \frac{\pi \langle S^z\rangle_n^4}{N'^2} \sum_{\mathbf{q}_\parallel \mathbf{p}_\parallel} [R_n^2(-\mathbf{k}_\parallel, \mathbf{p}_\parallel, \mathbf{q}_\parallel) + R_n^2(\mathbf{k}_\parallel - \mathbf{q}_\parallel + \mathbf{p}_\parallel, \mathbf{p}_\parallel, \mathbf{q}_\parallel)]\\ * \Big[\delta\Big(E^n_{\mathbf{p}_\parallel} - E^n_{\mathbf{q}_\parallel +} - \bar{\omega}^n_{\mathbf{k}_\parallel + \mathbf{p}_\parallel - \mathbf{q}_\parallel} + \bar{\omega}^n_{\mathbf{k}_\parallel}\Big)\\ - \delta\Big(E^n_{\mathbf{p}_\parallel} - E^n_{\mathbf{q}_\parallel} + \bar{\omega}^n_{\mathbf{q}_\parallel - \mathbf{k}_\parallel - \mathbf{p}_\parallel} + \bar{\omega}^n_{\mathbf{k}_\parallel}\Big)\Big]\delta_{q0}\delta_{p0}.\end{aligned} \tag{21}$$

It is seen that at $T = 0$ the phonon modes of the thin film are damped due to the spin–phonon interaction. Only the spin–phonon anharmonic terms contribute to γ^{ph} at $T = 0$.

The expression for the damping at $T \geqslant T_C$ is $\gamma^{ph} = \gamma^{ph-ph}$, because $\gamma^{sp} = 0$, i.e. only the phonon–phonon anharmonic terms contribute to the phonon damping in the vicinity of T_C and above it. This is because we have decoupled the longitudinal Green function, i.e. $\langle S_q^z S_q^z\rangle \rightarrow \langle S_0^z\rangle^2 \delta_{q0}$. If we take into account these correlation functions we would obtain a finite contribution from the spin–phonon interaction, i.e. $\gamma^{sp} \neq 0$.

The phonon damping $\gamma_{ph} = \gamma_{ph-ph} + \gamma_{sp-ph}$ for the phonon mode $\omega_0 = 100\,\mathrm{cm}^{-1}$ (D mode) for $CdCr_2Se_4$ is plotted in figure 6 as a function of temperature for various film thicknesses ($N = 16$ and 30 layers). The main signature of the spin–phonon contribution to the lifetime

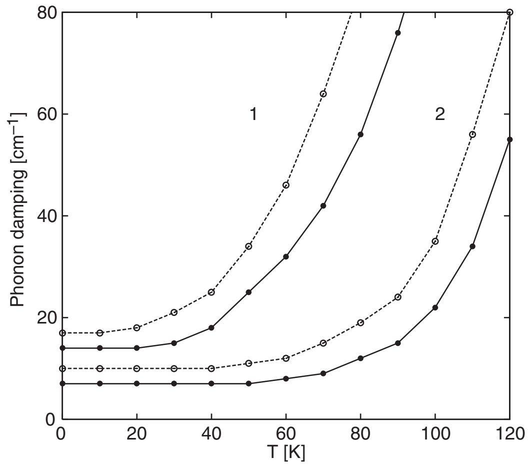

Figure 6. Temperature dependence of the phonon damping γ_{ph} for an sc ferroelectric film for the same parameters as in figure 1, and different film thicknesses: (1) $N = 16$, (2) 30 layers; dashed line—with, full line—without spin–phonon interaction.

broadening is the temperature dependence. The spin–phonon interaction enhances the phonon damping of the thin film. The damping increases near T_C, reaches a maximum, and then remains nearly constant. It can be seen that there are several differences between the thin films and the bulk behaviour. Thinner films have larger damping. For $N < 30$ layers we have

$$\Gamma_{TF}^{ph} > \Gamma_{B}^{ph}, \tag{22}$$

i.e. the damping is larger for thin films than that for the bulk, which is in agreement with the experimental data of Rohlsberger *et al* [26]. The anharmonicity at the surface is four to five times greater than in the bulk, so it is clear that the anharmonic terms play an important role in the lifetime broadening and must be taken into account if we want to obtain correct results for the damping effects at surfaces and in thin films.

6. Conclusions

Beyond the random phase approximation we get the renormalized spin-wave spectrum and phonon spectrum for an sc ferromagnetic semiconducting thin film. The spin–phonon coupling plays an important role. It decreases the spin-wave energy of the film. We show for the first time the importance and the influence of the spin–phonon interaction on the phonon spectrum of ferromagnetic semiconducting thin films. The phonon modes display a non-linear temperature dependence due to the influence of the spin ordering on the phonon modes. Only the anharmonic phonon–phonon interaction could not explain the temperature dependence below T_C. It has been found that the spin–phonon anharmonic terms play an important role at low temperatures, whereas the anharmonic phonon–phonon interaction is important at temperatures above T_C.

We have obtained that the spin-wave and phonon damping in ferromagnetic thin films is greater compared to the bulk case, due to different mechanisms which contribute additively to the damping, such as surface effects [12], electron–phonon interactions [18] and spin–phonon interactions, which are considered in this paper. It must be noted that the interactions of all three systems—the electrons, phonons and spins—must be considered in order to obtain correct results and to understand the experimental data in ferromagnetic semiconducting thin films.

Appendix A

Γ_n^{ss} is the damping part which arises from the spin–spin interaction:

$$\Gamma_n^{ss} = \frac{2\pi \langle S_n^z\rangle^2}{N'^2} \sum_{\mathbf{q}_\parallel \mathbf{p}_\parallel} v_n^2(\mathbf{k}_\parallel, \mathbf{q}_\parallel, \mathbf{p}_\parallel)\Big[\bar{n}^n_{\mathbf{p}_\parallel}(1 + \bar{n}^n_{\mathbf{k}_\parallel-\mathbf{q}_\parallel} + \bar{n}^n_{\mathbf{p}_\parallel+\mathbf{q}_\parallel}) - \bar{n}^n_{\mathbf{k}_\parallel-\mathbf{q}_\parallel}\bar{n}^n_{\mathbf{p}_\parallel+\mathbf{q}_\parallel}\Big]$$
$$\times\, \delta(E^n_{\mathbf{p}_\parallel+\mathbf{q}_\parallel} + E^n_{\mathbf{k}_\parallel-\mathbf{q}_\parallel} - E^n_{\mathbf{p}_\parallel} - E^n_{\mathbf{k}_\parallel}), \tag{23}$$

where

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

Γ_n^{sd} is the damping which arises from the interaction between the ferromagnetically ordered and the conduction band electrons:

$$\Gamma_n^{sd} = \frac{2\pi I_n^2\langle S_n^z\rangle}{N'^3} \sum_{\mathbf{q}_\parallel \mathbf{p}_\parallel \mathbf{r}_\parallel} \Big[(\bar{n}^n_{\mathbf{p}_\parallel} - \bar{n}^n_{\mathbf{p}_\parallel+\mathbf{k}_\parallel+\mathbf{q}_\parallel})\bar{m}_{\mathbf{q}_\parallel+\mathbf{r}_\parallel+}(1 - \bar{m}_{\mathbf{r}_\parallel-}),$$
$$+ \bar{n}^n_{\mathbf{p}_\parallel+\mathbf{k}_\parallel+\mathbf{q}_\parallel}(1 + \bar{n}^n_{\mathbf{p}_\parallel})(\bar{m}_{\mathbf{q}_\parallel+\mathbf{r}_\parallel+} - \bar{m}_{\mathbf{r}_\parallel-})\Big]$$
$$* \,\delta(E^n_{\mathbf{p}_\parallel+\mathbf{k}_\parallel+\mathbf{q}_\parallel} - E^n_{\mathbf{p}_\parallel} + \epsilon^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel+} - \epsilon^n_{\mathbf{r}_\parallel-} - E^n_{\mathbf{k}_\parallel})$$
$$+ \frac{\pi I_n^2}{4N'^2} \sum_{\mathbf{q}_\parallel \mathbf{p}_\parallel \sigma} \Big[\bar{m}_{\mathbf{p}_\parallel+\mathbf{q}_\parallel\sigma}(1 - \bar{m}_{\mathbf{p}_\parallel\sigma}) + \bar{n}^n_{\mathbf{k}_\parallel-\mathbf{q}_\parallel}(\bar{m}_{\mathbf{p}_\parallel+\mathbf{q}_\parallel\sigma} - \bar{m}_{\mathbf{p}_\parallel\sigma})\Big]$$
$$* \,\delta(E^n_{\mathbf{k}_\parallel-\mathbf{q}_\parallel} + \epsilon^n_{\mathbf{p}_\parallel+\mathbf{q}_\parallel\sigma} - \epsilon^n_{\mathbf{p}_\parallel\sigma} - E^n_{\mathbf{k}_\parallel})$$
$$+ \frac{\pi I_n^2\langle S_n^z\rangle}{2N'} \sum_{\mathbf{q}_\parallel}(\bar{m}_{\mathbf{q}_\parallel\mathbf{k}_\parallel+} - \bar{m}_{\mathbf{q}_\parallel-})\delta(\epsilon^n_{\mathbf{q}_\parallel-\mathbf{k}_\parallel+} - \epsilon^n_{\mathbf{q}_\parallel-} - E^n_{\mathbf{k}_\parallel}). \tag{24}$$

Γ_n^{sp} is the damping due to the spin–phonon interaction:

$$\Gamma_n^{sp} = \frac{2\pi\langle S_n^z\rangle^2}{N'^3} \sum_{\mathbf{q}_\parallel \mathbf{p}_\parallel \mathbf{r}_\parallel} \Big(F_n^2(\mathbf{k}_\parallel, \mathbf{q}_\parallel, \mathbf{p}_\parallel, \mathbf{r}_\parallel)\Big[\bar{n}^n_{\mathbf{r}_\parallel}(1 + \bar{n}^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel} + \bar{n}^n_{\mathbf{p}_\parallel}) - \bar{n}^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel}\bar{n}^n_{\mathbf{p}_\parallel}\Big]$$
$$* \Big[(1 + \bar{N}_{\mathbf{k}_\parallel-\mathbf{p}_\parallel-\mathbf{q}_\parallel})\delta(E^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel} - E^n_{\mathbf{r}_\parallel} + E^n_{\mathbf{p}_\parallel} - \bar{\omega}_{\mathbf{k}_\parallel-\mathbf{p}_\parallel-\mathbf{q}_\parallel} - E^n_{\mathbf{k}_\parallel})$$
$$+ (\bar{N}_{\mathbf{k}_\parallel-\mathbf{p}_\parallel-\mathbf{q}_\parallel})\delta(E^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel} - E^n_{\mathbf{r}_\parallel} + E^n_{\mathbf{p}_\parallel} + \bar{\omega}_{\mathbf{k}_\parallel-\mathbf{p}_\parallel-\mathbf{q}_\parallel} - E^n_{\mathbf{k}_\parallel})\Big]$$
$$+ F_n^2(\mathbf{k}_\parallel, \mathbf{q}_\parallel, \mathbf{p}_\parallel, \mathbf{r}_\parallel)(1 + \bar{n}^n_{\mathbf{p}_\parallel})(1 + \bar{n}^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel})\bar{n}^n_{\mathbf{r}_\parallel}\Big[\delta(E^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel} - E^n_{\mathbf{r}_\parallel}$$
$$+ E^n_{\mathbf{p}_\parallel} - \bar{\omega}_{\mathbf{k}_\parallel-\mathbf{p}_\parallel-\mathbf{q}_\parallel} - E^n_{\mathbf{k}_\parallel}) - \delta(E^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel} - E^n_{\mathbf{r}_\parallel} + E^n_{\mathbf{p}_\parallel} + \bar{\omega}_{\mathbf{k}_\parallel-\mathbf{p}_\parallel-\mathbf{q}_\parallel} - E^n_{\mathbf{k}_\parallel})\Big]\Big)$$
$$+ \frac{\pi}{4N}\sum_{\mathbf{q}_\parallel} F_n^2(\mathbf{k}_\parallel, \mathbf{q}_\parallel)\Big[(1 + \bar{N}^n_{\mathbf{q}_\parallel-\mathbf{k}_\parallel} + \bar{n}^n_{\mathbf{q}_\parallel})\delta(E^n_{\mathbf{q}_\parallel} - \bar{\omega}_{\mathbf{q}_\parallel-\mathbf{k}_\parallel} - E^n_{\mathbf{k}_\parallel})$$
$$+ (\bar{N}^n_{\mathbf{q}_\parallel-\mathbf{k}_\parallel} - \bar{n}^n_{\mathbf{q}_\parallel})\delta(E^n_{\mathbf{q}_\parallel} + \bar{\omega}_{\mathbf{q}_\parallel-\mathbf{k}_\parallel} - E^n_{\mathbf{k}_\parallel})\Big]$$
$$+ \frac{\pi}{2N'^3}\sum_{\mathbf{q}_\parallel \mathbf{p}_\parallel \mathbf{r}_\parallel}\Big(R_n^2(\mathbf{q}_\parallel, \mathbf{p}_\parallel, \mathbf{r}_\parallel)\Big[\Big[(1 + \bar{N}^n_{\mathbf{p}_\parallel-\mathbf{q}_\parallel-\mathbf{r}_\parallel})(1 + \bar{N}^n_{\mathbf{r}_\parallel}) + \bar{N}^n_{\mathbf{r}_\parallel}\bar{n}^n_{\mathbf{k}_\parallel+\mathbf{q}_\parallel-\mathbf{p}_\parallel}\Big]$$
$$* \,\delta(E^n_{\mathbf{k}_\parallel+\mathbf{q}_\parallel-\mathbf{p}_\parallel} + \bar{\omega}_{\mathbf{r}_\parallel} + \bar{\omega}_{\mathbf{p}_\parallel-\mathbf{q}_\parallel-\mathbf{r}_\parallel} + E^n_{\mathbf{k}_\parallel})$$
$$+ \Big[\bar{N}^n_{\mathbf{r}_\parallel}\bar{N}^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel-\mathbf{p}_\parallel} - (1 + \bar{N}^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel-\mathbf{p}_\parallel} + \bar{N}^n_{\mathbf{r}_\parallel})\bar{n}^n_{\mathbf{k}_\parallel+\mathbf{q}_\parallel-\mathbf{p}_\parallel}\Big]$$
$$* \,\delta(E^n_{\mathbf{k}_\parallel+\mathbf{q}_\parallel-\mathbf{p}_\parallel} - \bar{\omega}_{\mathbf{r}_\parallel} - \bar{\omega}_{\mathbf{p}_\parallel-\mathbf{q}_\parallel-\mathbf{r}_\parallel} + E^n_{\mathbf{k}_\parallel})\Big]$$

$$
\begin{aligned}
&+ R_n(\mathbf{q}_\parallel, \mathbf{p}_\parallel, \mathbf{r}_\parallel) R_n(\mathbf{p}_\parallel - \mathbf{q}_\parallel - \mathbf{r}_\parallel, \mathbf{p}_\parallel, \mathbf{q}_\parallel)\Big[(1 + \bar{N}^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel-\mathbf{p}_\parallel})(1 + \bar{n}^n_{\mathbf{k}_\parallel+\mathbf{q}_\parallel-\mathbf{p}_\parallel}) \\
&* \delta(E^n_{\mathbf{k}_\parallel+\mathbf{q}_\parallel-\mathbf{p}_\parallel} + \bar{\omega}_{\mathbf{r}_\parallel} + \bar{\omega}_{\mathbf{p}_\parallel-\mathbf{q}_\parallel-\mathbf{r}_\parallel} + E^n_{\mathbf{k}_\parallel}) \\
&- \Big[\bar{N}^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel-\mathbf{p}_\parallel} + (1 + \bar{N}^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel-\mathbf{p}_\parallel})\bar{n}^n_{\mathbf{k}_\parallel+\mathbf{q}_\parallel-\mathbf{p}_\parallel}\Big] \\
&* \delta(E^n_{\mathbf{k}_\parallel+\mathbf{q}_\parallel-\mathbf{p}_\parallel} - \bar{\omega}_{\mathbf{r}_\parallel} - \bar{\omega}_{\mathbf{p}_\parallel-\mathbf{q}_\parallel-\mathbf{r}_\parallel} + E^n_{\mathbf{k}_\parallel})\Big]\Big) \\
&+ \frac{\pi}{2N'^3} \sum_{\mathbf{q}_\parallel\mathbf{p}_\parallel\mathbf{r}_\parallel} \Big(R^2_n(\mathbf{q}_\parallel, \mathbf{p}_\parallel, \mathbf{r}_\parallel)\Big[(1 + \bar{N}^n_{\mathbf{p}_\parallel-\mathbf{q}_\parallel-\mathbf{r}_\parallel})\bar{N}^n_{\mathbf{r}_\parallel} \\
&+ (\bar{N}^n_{\mathbf{p}_\parallel-\mathbf{q}_\parallel-\mathbf{r}_\parallel} - \bar{N}^n_{\mathbf{r}_\parallel})\bar{n}^n_{\mathbf{k}_\parallel+\mathbf{q}_\parallel-\mathbf{p}_\parallel}\Big] \\
&* \Big[\delta(E^n_{\mathbf{k}_\parallel+\mathbf{q}_\parallel-\mathbf{p}_\parallel} + \bar{\omega}_{\mathbf{r}_\parallel} - \bar{\omega}_{\mathbf{p}_\parallel-\mathbf{q}_\parallel-\mathbf{r}_\parallel} + E^n_{\mathbf{k}_\parallel}) \\
&+ \delta(E^n_{\mathbf{k}_\parallel+\mathbf{q}_\parallel-\mathbf{p}_\parallel} - \bar{\omega}_{\mathbf{r}_\parallel} + \bar{\omega}_{\mathbf{p}_\parallel-\mathbf{q}_\parallel-\mathbf{r}_\parallel} + E^n_{\mathbf{k}_\parallel})\Big]\Big) \\
&+ \frac{4\pi\langle S^z_n\rangle^2}{N'^4} \sum_{\mathbf{q}_\parallel\mathbf{p}_\parallel\mathbf{r}_\parallel\nu_\parallel} \Big(R^2_n(\mathbf{k}_\parallel, \mathbf{q}_\parallel, \mathbf{p}_\parallel, \mathbf{r}_\parallel, \nu_\parallel)\Big[\bar{n}^n_{\mathbf{r}_\parallel}(1 + \bar{n}^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel} + \bar{n}^n_{\mathbf{p}_\parallel}) - \bar{n}^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel}\bar{n}^n_{\mathbf{p}_\parallel}\Big] \\
&* \Big[(1 + \bar{N}_{\mathbf{k}_\parallel-\mathbf{p}_\parallel-\mathbf{q}_\parallel-\nu_\parallel} + \bar{N}^n_{\nu_\parallel})\delta(E^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel} - E^n_{\mathbf{r}_\parallel} + E^n_{\mathbf{p}_\parallel} + \bar{\omega}_{\nu_\parallel} + \bar{\omega}_{\mathbf{k}_\parallel-\mathbf{p}_\parallel-\mathbf{q}_\parallel-\nu_\parallel} - E^n_{\mathbf{k}_\parallel}) \\
&+ (\bar{N}_{\mathbf{k}_\parallel-\mathbf{p}_\parallel-\mathbf{q}_\parallel-\nu_\parallel} + \bar{N}^n_{\nu_\parallel})\delta(E^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel} - E^n_{\mathbf{r}_\parallel} + E^n_{\mathbf{p}_\parallel} - \bar{\omega}_{\nu_\parallel} - \bar{\omega}_{\mathbf{k}_\parallel-\mathbf{p}_\parallel-\mathbf{q}_\parallel-\nu_\parallel} - E^n_{\mathbf{k}_\parallel})\Big] \\
&+ R^2_n(\mathbf{k}_\parallel, \mathbf{q}_\parallel, \mathbf{p}_\parallel, \mathbf{r}_\parallel, \nu_\parallel)(1 + \bar{n}^n_{\mathbf{p}_\parallel})(1 + \bar{n}^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel})\bar{n}^n_{\mathbf{r}_\parallel}(\bar{N}_{\mathbf{k}_\parallel-\mathbf{p}_\parallel-\mathbf{q}_\parallel-\nu_\parallel} + \bar{N}^n_{\nu_\parallel}) \\
&* \Big[\delta(E^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel} - E^n_{\mathbf{r}_\parallel} + E^n_{\mathbf{p}_\parallel} + \bar{\omega}_{\nu_\parallel} - \bar{\omega}_{\mathbf{k}_\parallel-\mathbf{p}_\parallel-\mathbf{q}_\parallel-\nu_\parallel} - E^n_{\mathbf{k}_\parallel}) \\
&- \delta(E^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel} - E^n_{\mathbf{r}_\parallel} + E^n_{\mathbf{p}_\parallel} - \bar{\omega}_{\nu_\parallel} + \bar{\omega}_{\mathbf{k}_\parallel-\mathbf{p}_\parallel-\mathbf{q}_\parallel-\nu_\parallel} - E^n_{\mathbf{k}_\parallel})\Big]\Big) \\
&+ \frac{4\pi\langle S^z_n\rangle^2}{N'^4} \sum_{\mathbf{q}_\parallel\mathbf{p}_\parallel\mathbf{r}_\parallel\nu_\parallel} R^2_n(\mathbf{k}_\parallel, \mathbf{q}_\parallel, \mathbf{p}_\parallel, \mathbf{r}_\parallel, \nu_\parallel)\Big[\bar{n}^n_{\mathbf{r}_\parallel}(1 + \bar{n}^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel} + \bar{n}^n_{\mathbf{p}_\parallel}) - \bar{n}^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel}\bar{n}^n_{\mathbf{p}_\parallel}\Big] \\
&* (1 + \bar{N}_{\mathbf{k}_\parallel-\mathbf{p}_\parallel-\mathbf{q}_\parallel-\nu_\parallel})\Big[\delta(E^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel} - E^n_{\mathbf{r}_\parallel} + E^n_{\mathbf{p}_\parallel} + \bar{\omega}_{\nu_\parallel} + \bar{\omega}_{\mathbf{k}_\parallel-\mathbf{p}_\parallel-\mathbf{q}_\parallel-\nu_\parallel} - E^n_{\mathbf{k}_\parallel}) \\
&- \delta(E^n_{\mathbf{q}_\parallel+\mathbf{r}_\parallel} - E^n_{\mathbf{r}_\parallel} + E^n_{\mathbf{p}_\parallel} - \bar{\omega}_{\nu_\parallel} - \bar{\omega}_{\mathbf{k}_\parallel-\mathbf{p}_\parallel-\mathbf{q}_\parallel-\nu_\parallel} - E^n_{\mathbf{k}_\parallel})\Big].
\end{aligned} \tag{25}
$$

Appendix B

γ^{sp}_n is the damping part which comes from the spin–phonon interaction:

$$
\begin{aligned}
\gamma^{\mathrm{sp}}_n = {}& \frac{4\pi\langle S^z\rangle^2_n}{N'} \sum_{\mathbf{q}_\parallel} F^2_n(\mathbf{q}_\parallel, \mathbf{q}_\parallel - \mathbf{k}_\parallel)(\bar{n}^n_{\mathbf{q}_\parallel} - \bar{n}^n_{\mathbf{q}_\parallel-\mathbf{k}_\parallel})\delta\Big(E^n_{\mathbf{q}_\parallel-\mathbf{k}_\parallel} - E^n_{\mathbf{q}_\parallel} - \bar{\omega}^n_{\mathbf{k}_\parallel}\Big) \\
&+ \frac{4\pi\langle S^z\rangle^2_n}{N'^2} \sum_{\mathbf{q}_\parallel,\mathbf{p}_\parallel} \Big(R^2_n(-\mathbf{k}_\parallel, \mathbf{p}_\parallel, \mathbf{q}_\parallel)(\bar{n}^n_{\mathbf{q}_\parallel} - \bar{n}^n_{\mathbf{p}_\parallel})\Big[(1 + \bar{N}^n_{\mathbf{k}_\parallel+\mathbf{p}_\parallel-\mathbf{q}_\parallel}) \\
&* \delta\Big(E^n_{\mathbf{p}_\parallel} - E^n_{\mathbf{q}_\parallel} - \bar{\omega}^n_{\mathbf{k}_\parallel+\mathbf{p}_\parallel-\mathbf{q}_\parallel} + \bar{\omega}^n_{\mathbf{k}_\parallel}\Big) + \bar{N}^n_{\mathbf{q}_\parallel-\mathbf{k}_\parallel-\mathbf{p}_\parallel} \\
&* \delta\Big(E^n_{\mathbf{p}_\parallel} - E^n_{\mathbf{q}_\parallel} + \bar{\omega}^n_{\mathbf{q}_\parallel-\mathbf{k}_\parallel-\mathbf{p}_\parallel} + \bar{\omega}^n_{\mathbf{k}_\parallel}\Big)\Big] \\
&+ \Big[R^2_n(-\mathbf{k}_\parallel, \mathbf{p}_\parallel, \mathbf{q}_\parallel) + R^2_n(-\mathbf{k}_\parallel - \mathbf{q}_\parallel + \mathbf{p}_\parallel, \mathbf{p}_\parallel, \mathbf{q}_\parallel)\Big]\bar{n}^n_{\mathbf{q}_\parallel}(1 + \bar{n}^n_{\mathbf{p}_\parallel}) \\
&* \Big[\delta\Big(E^n_{\mathbf{p}_\parallel} - E^n_{\mathbf{q}_\parallel+} - \bar{\omega}^n_{\mathbf{k}_\parallel+\mathbf{p}_\parallel-\mathbf{q}_\parallel} + \bar{\omega}^n_{\mathbf{k}_\parallel}\Big) - \delta\Big(E^n_{\mathbf{p}_\parallel} - E^n_{\mathbf{q}_\parallel} + \bar{\omega}^n_{\mathbf{q}_\parallel-\mathbf{k}_\parallel-\mathbf{p}_\parallel} + \bar{\omega}^n_{\mathbf{k}_\parallel}\Big)\Big]\Big)
\end{aligned}
$$

$$+ \frac{\pi}{N'^2} \sum_{\mathbf{q}_\parallel \mathbf{p}_\parallel} \Big[R_n^2(-\mathbf{k}_\parallel, \mathbf{p}_\parallel, \mathbf{q}_\parallel) + R_n^2(\mathbf{k}_\parallel - \mathbf{q}_\parallel + \mathbf{p}_\parallel, \mathbf{p}_\parallel, \mathbf{q}_\parallel) \Big] \langle S^z \rangle_n^4 \delta_{q0} \delta_{p0}$$
$$* \Big[\delta\Big(E^n_{\mathbf{p}_\parallel} - E^n_{\mathbf{q}_\parallel +} - \bar{\omega}^n_{\mathbf{k}_\parallel + \mathbf{p}_\parallel - \mathbf{q}_\parallel} + \bar{\omega}^n_{\mathbf{k}_\parallel} \Big) - \delta\Big(E^n_{\mathbf{p}_\parallel} - E^n_{\mathbf{q}_\parallel} + \bar{\omega}^n_{\mathbf{q}_\parallel - \mathbf{k}_\parallel - \mathbf{p}_\parallel} + \bar{\omega}^n_{\mathbf{k}_\parallel} \Big) \Big]. \quad (26)$$

$\gamma_n^{\mathrm{ph-ph}}$ is the phonon damping due to the phonon–phonon anharmonic interaction:

$$\gamma_n^{\mathrm{ph-ph}} = \frac{3\pi}{N'} \sum_{\mathbf{q}_\parallel} [B_n^2(\mathbf{q}_\parallel, -\mathbf{k}_\parallel, \mathbf{k}_\parallel - \mathbf{q}_\parallel) + B_n^2(\mathbf{q}_\parallel, \mathbf{k}_\parallel - \mathbf{q}_\parallel, -\mathbf{k}_\parallel)]$$
$$* (\bar{N}^n_{\mathbf{q}_\parallel} - \bar{N}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel})[\delta(\bar{\omega}^n_{\mathbf{k}_\parallel} - \bar{\omega}^n_{\mathbf{q}_\parallel} - \bar{\omega}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel}) + \delta(\bar{\omega}^n_{\mathbf{k}_\parallel} - \bar{\omega}^n_{\mathbf{q}_\parallel} + \bar{\omega}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel})]$$
$$+ \frac{8\pi}{N'} \sum_{\mathbf{q}_\parallel} \Big[A_n^2(\mathbf{q}_\parallel, -\mathbf{k}_\parallel, \mathbf{p}_\parallel, \mathbf{k}_\parallel - \mathbf{q}_\parallel - \mathbf{p}_\parallel) + A_n^2(\mathbf{q}_\parallel, \mathbf{p}_\parallel, -\mathbf{k}_\parallel, \mathbf{k}_\parallel - \mathbf{q}_\parallel - \mathbf{p}_\parallel) \Big]$$
$$* \Big[\bar{N}^n_{\mathbf{p}_\parallel} \Big(1 + \bar{N}^n_{\mathbf{q}_\parallel} + \bar{N}^n_{\mathbf{p}_\parallel + \mathbf{k}_\parallel - \mathbf{q}_\parallel} \Big) - \bar{N}^n_{\mathbf{q}_\parallel} \bar{N}^n_{\mathbf{p}_\parallel + \mathbf{k}_\parallel - \mathbf{q}_\parallel} \Big]$$
$$* \delta\Big(\bar{\omega}^n_{\mathbf{k}_\parallel} - \bar{\omega}^n_{\mathbf{q}_\parallel} + \bar{\omega}^n_{\mathbf{p}_\parallel} - \bar{\omega}^n_{\mathbf{k}_\parallel + \mathbf{p}_\parallel - \mathbf{q}_\parallel} \Big). \quad (27)$$

References

[1] Rezende S M, Azevedo A, Lucena M A and de Aguiar F M 2001 *Phys. Rev.* B **63** 214418
[2] McMichael R D, Stiles M D, Chen P J and Egelhoff W F Jr 1998 *J. Appl. Phys.* **83** 7037
[3] Fermin J R, Azevedo A, de Aguiar F M, Li B and Rezende S M 1999 *J. Appl. Phys.* **85** 7316
[4] Azevedo A, Olivera A B, de Aguiar F M and Rezende S M 2000 *Phys. Rev.* B **62** 5331
[5] Heinrich B, Urban R and Woltersdorf G 2002 *J. Appl. Phys.* **91** 7523
[6] Arias R and Mills D L 1999 *Phys. Rev.* B **60** 7395
[7] Dobin A Yu and Victora R H 2003 *Phys. Rev. Lett.* **90** 176203
[8] Costa A T, Muniz R B and Mills D L 2003 *Phys. Rev.* B **68** 224435
[9] Costa A T Jr, Muniz R B and Mills D L 2004 *Phys. Rev.* B **70** 054406
[10] Gopalan S and Cottam M G 1990 *Phys. Rev.* B **42** 10311
[11] Mills D L 2003 *Phys. Rev.* B **68** 014419
[12] Wesselinowa J M, Iliev L L and Nolting W 1999 *Phys. Status Solidi* b **214** 165
[13] Wesselinowa J M 2003 *J. Magn. Magn. Mater.* **262** 264
[14] Quinn J J 1958 *Phys. Rev.* **112** 812
Quinn J J 1962 *Phys. Rev.* **126** 1453
[15] McDougall B A, Balasubramanian T and Jensen E 1995 *Phys. Rev.* B **51** 13891
[16] Hellsing B, Eiguren A and Chulkov E V 2002 *J. Phys.: Condens. Matter* **14** 5959
[17] Allen P B 2001 *Phys. Rev.* B **63** 214410
[18] Wesselinowa J M 2005 *J. Phys.: Condens. Matter* **17** 507
[19] Haas C and Callen H B 1963 *Ferromagnetic Relaxation and Resonance Line Width* vol 1 *Magnetism* (New York: Academic)
[20] Iliev M N, Litvinchuk A P, Lee H-G, Chu C W, Barry A and Coey J M D 1999 *Phys. Rev.* B **60** 33
[21] McMichael R D and Kunz A 2002 *J. Appl. Phys.* **91** 8650
[22] Booth C H, Bridges F, Kwei G H, Lawrence J M, Cornelius A L and Neumeier J J 1998 *Phys. Rev. Lett.* **80** 853
[23] Rehbein A, Wegner D, Kaindl G and Bauer A 2003 *Phys. Rev.* B **67** 033403
[24] Chang Y M, Xu L and Tom H W K 1999 *Phys. Rev.* B **59** 12220
[25] Watanabe K, Dimitrov D T, Takagi N and Matsumoto Y 2002 *Phys. Rev.* B **65** 235328
[26] Rohlsberger R, Sturhahn W, Toellner T S, Quast K W, Hession P, Hu M, Sutter J and Alp E E 1999 *J. Appl. Phys.* **86** 584
[27] Baddorf A P and Plummer E W 1991 *Phys. Rev. Lett.* **66** 2770
[28] Rahman T S, Spangler J D and Al-Rawi A 2002 *J. Phys.: Condens. Matter* **14** 5903
[29] Tserkovnikov Yu 1971 *Theor. Math. Phys.* **7** 250
[30] Wesselinowa J M and Apostolov A T 1996 *J. Phys.: Condens. Matter* **8** 473
[31] Ma S K S, de Wette F W and Alldredge G P 1978 *Surf. Sci.* **78** 598
[32] Jayanthi C S and Tosatti E 1985 *Phys. Rev.* B **31** 3456