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2005 J. Phys.: Condens. Matter 17 507

(http://iopscience.iop.org/0953-8984/17/3/010)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **17** (2005) 507–517 doi:10.1088/0953-8984/17/3/010

Electron–phonon interaction in ferromagnetic semiconducting thin films

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Received 16 November 2004, in final form 16 December 2004
Published 7 January 2005
Online at stacks.iop.org/JPhysCM/17/507

Abstract

The effect of electron–phonon interaction on the temperature dependence of the electronic spectrum in thin ferromagnetic semiconducting films is studied using the s–d interaction model and a Green function formalism beyond the random phase approximation. We obtain three temperature regions with different contributions of the damping terms due to the s–d, electron–electron and electron–phonon interactions. The phonon spectrum is discussed, too. Additional phonon damping and phonon frequency shift arise when the electron–phonon interaction is properly included. The phonon linewidth may be obtained from the cubic terms of the anharmonic phonon–phonon interaction, while the calculation of the renormalized phonon frequency of the experimentally obtained frequency shift required inclusion of the quartic term in the anharmonic interaction. The frequencies in thin ferromagnetic semiconducting films are smaller and the damping effects larger in comparison with the bulk case.

1. Introduction

Magnetic thin films are not only useful for practical applications such as in magneto-optical recording but also important in the study of inhomogeneous spin systems in low dimensions. The properties of magnetic films depend on the film thickness. The Curie temperature T_C, the value of the magnetization M, the orientation of the magnetic moments etc can depend drastically on the thickness and structure of the film [1–3]. Experiments [4] have shown that in the general case the Curie temperature of thin films is lower than the bulk Curie temperature and, as the thickness of a thin film decreases, its Curie temperature also decreases. In some special cases, T_C for thin films is higher than the bulk Curie temperature, for example in Gd [5], V [6] and epitaxial films [7].

Whereas the magnetic properties of thin magnetic films have been extensively theoretically investigated, this is not the case for ferromagnetic semiconducting thin films. The magnetic properties of thin films containing itinerant electrons interacting with localized spins and the problem of inter-layer coupling have been considered by Urbaniak-Kucharczyk [8]. The magnetic properties of thin ferromagnetic and antiferromagnetic itinerant-electron films are

investigated within the single-band Hubbard model by Wu and Nolting [9]. Gopalan and Cottam [10] have used the s–d interaction model to study the bulk and surface magnetic excitations of a semi-infinite ferromagnetic semiconductor for low temperatures. 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 function formalism by Wesselinowa *et al* [11]. The dynamical properties of thin ferromagnetic semiconducting films are obtained by Wesselinowa [12]. 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 [13], electron–phonon [14, 15] and electron–magnon interactions [16]. Despite attracting considerable interest for half a century since the pioneering work by Fröhlich [17], the problem of electron–phonon interaction is still far from being solved, especially for thin films. The temperature dependence of the lifetime broadening of the Gd(0001) surface state is studied using scanning tunnelling spectroscopy [18]. The observed increase of the linewidth with temperature is attributed to enhanced electron–phonon scattering. Coherent surface phonons at a GaAs surface have been investigated by means of time-resolved second-harmonic generation [19, 20]. 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. Generally, the anharmonicity of surface phonons modes is considered to be greater than that of bulk phonon modes. Baddorf and Plummer [21] have revealed that the anharmonicity for the motion normal to the surface on a Cu(110) surface is 4–5 times greater than that in bulk copper. Valla *et al* [22] reported measurements of the electron–phonon coupling in ultrathin silver films deposited on a V(100) surface, obtained from the temperature dependence of the widths of angle-resolved photoelectron spectroscopy peaks from quantum well states in the film. A strong oscillatory variation in coupling strength is observed as a function of film thickness.

Theoretical studies of surface phonon linewidths of Ag, Cu and Al are presented by Rahman *et al* [23]. The electron–phonon coupling at metal surfaces is investigated by Hellsing *et al* [15]. They have demonstrated that it is possible to understand experimental data concerning the electron–phonon-induced lifetime broadening of surface states reasonably well by taking into account bulk and surface electron and phonon states. The purpose of the present paper is to investigate the electron–phonon interaction in ferromagnetic semiconducting thin films on the basis of the s–d interaction model.

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 $n = 1, \ldots, N$, where the $n = 1$ and N layers 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:

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

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

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

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 originating from 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 $J_{ij} = J_s$. Otherwise, the interaction in the bulk material is written as J, which is for simplicity assumed to be the same as that 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. Similar notation is used for all parameters in equations (2)–(7).

H_E represents the usual Hamiltonian of the conduction band electrons,

$$H_E = \sum_{l,\delta,\sigma} t_{l,l+\delta} c^+_{l\sigma} c_{l+\delta,\sigma}, \tag{3}$$

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

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

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

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

H_{EE} is the electron–electron interaction term:

$$H_{EE} = \tfrac{1}{2} \sum_{\mathbf{q}\mathbf{k}'\mathbf{k}''\sigma\sigma'} v(\mathbf{q}) c^+_{\mathbf{k}''-\mathbf{q}\sigma} c^+_{\mathbf{k}'+\mathbf{q}\sigma'} c_{\mathbf{k}'\sigma'} c_{\mathbf{k}''\sigma}; \tag{5}$$

$v(\mathbf{q}) = 4\pi e^2/\kappa \mathbf{q}^2$ is the Coulomb interaction, where e is the electron charge and κ is the dielectric constant.

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

$$H_P = \frac{1}{2!} \sum_{\mathbf{q}} (P_{\mathbf{q}} P_{-\mathbf{q}} + \omega^2_{\mathbf{q}} Q_{\mathbf{q}} Q_{-\mathbf{q}}) + \frac{1}{3!} \sum_{\mathbf{q}\mathbf{q}_1} B(\mathbf{q}, \mathbf{q}_1) Q_{\mathbf{q}} Q_{-\mathbf{q}_1} Q_{\mathbf{q}_1-\mathbf{q}} + \frac{1}{4!} \sum_{\mathbf{q},\mathbf{q}_1,\mathbf{q}_2} A(\mathbf{q}, \mathbf{q}_1, \mathbf{q}_2) Q_{\mathbf{q}_1} Q_{\mathbf{q}_2} Q_{-\mathbf{q}-\mathbf{q}_2} Q_{-\mathbf{q}_1+\mathbf{q}}, \tag{6}$$

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: $Q_{\mathbf{q}\lambda} = (2\omega_{\mathbf{q}\lambda})^{-1/2}(a_{\mathbf{q}\lambda} + a^+_{-\mathbf{q}\lambda})$, $P_q = \mathrm{i}(\omega_{\mathbf{q}\lambda}/2)^{1/2}(a^+_{\mathbf{q}\lambda} - a_{-\mathbf{q}\lambda})$.

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

$$H_{EP} = -\tfrac{1}{2} \sum_{\mathbf{q}\mathbf{p}\sigma} F(\mathbf{q}, \mathbf{p}) Q_{\mathbf{p}-\mathbf{q}} \rho_{\mathbf{q}\sigma} - \tfrac{1}{4} \sum_{\mathbf{q}\mathbf{p}\nu\sigma} R(\mathbf{q}, \mathbf{p}, \nu) Q_\nu Q_{\mathbf{p}-\mathbf{q}-\nu} \rho_{\mathbf{q}\sigma} + \text{h.c.} \tag{7}$$

with $\rho_{\mathbf{q}\sigma} = \sum_{\mathbf{k}} c^+_{\mathbf{k}\sigma} c_{\mathbf{k}+\mathbf{q}\sigma}$.

3. The one-electron Green function

To study the electronic excitations of the film we introduce the one-electron Green function $g_{ij\sigma}(E) = \langle\langle c_{i\sigma}; c^+_{j\sigma}\rangle\rangle$. On introducing the two-dimensional Fourier transform $g_{n_in_j}(\mathbf{k}_\parallel, E)$, one has the following form:

$$\langle\langle c^+_{i\sigma}; c^-_{j\sigma}\rangle\rangle = \frac{1}{N'}\sum_{\mathbf{k}_\parallel}\exp(\mathrm{i}\mathbf{k}_\parallel(\mathbf{r}_i - \mathbf{r}_j))g_{n_in_j}(\mathbf{k}_\parallel, E), \tag{8}$$

where N' is the number of sites in any of the lattice planes, $\mathbf{r}_i$ represents the position vectors of site i, $n = 1, \ldots, N$ denotes the layer ordering number beginning with one surface ($n = 1$) and terminating with the other surface ($n = N$), $\mathbf{k}_\parallel = (k_x, k_y)$ is a two-dimensional wavevector parallel to the surface. The summation is taken over the Brillouin zone.

We assume for simplicity only nearest-neighbour exchange interactions and write all interaction constants for the surface layers ($n = 1, N$) with index 's' and all others without an index. For the approximate calculation of the Green function (8) we use a method proposed by Tserkovnikov [27], which is appropriate for spin problems. It goes beyond the random phase approximation (RPA) taking into account the correlation functions $\bar{N}_{\mathbf{q}_\parallel} = \langle S^+_{\mathbf{q}_\parallel} S^-_{\mathbf{q}_\parallel}\rangle$, $\bar{n}_{\mathbf{q}_\parallel\sigma} = \langle c^+_{\mathbf{q}_\parallel\sigma} c_{\mathbf{q}_\parallel\sigma}\rangle$ and $\bar{m}_{\mathbf{q}_\parallel} = \langle a^+_{\mathbf{q}_\parallel} a_{\mathbf{q}_\parallel}\rangle$, and the damping effects. As a result the equation of motion for the Green function (8) of the ferromagnetic semiconducting film for $T \leqslant T_c$ has the following matrix form:

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

where $\mathbf{I}$ is the unit matrix and

$$\mathbf{L}(\mathbf{E}) = \begin{pmatrix} E - L^\sigma_1 + \mathrm{i}\Gamma^\sigma_1 & -k & 0 & 0 & 0 & 0 & \ldots \\ -k & E - L^\sigma_2 + \mathrm{i}\Gamma^\sigma_2 & -k & 0 & 0 & 0 & \ldots \\ 0 & -k & E - L^\sigma_3 + \mathrm{i}\Gamma^\sigma_3 & -k & 0 & 0 & \ldots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \ddots \\ 0 & 0 & 0 & 0 & 0 & -k & E - L^\sigma_N + \mathrm{i}\Gamma^\sigma_N \end{pmatrix}$$

with

$k = t$,

$$L^+_n = -\tfrac{1}{2}(I_n\langle S^z\rangle_n) + 4t_n\gamma(\mathbf{k}_\parallel) + \sum_{\mathbf{q}_\parallel}\big(v_n(o) - v_n(\mathbf{k}_\parallel - \mathbf{q}_\parallel)\big)\bar{n}^n_{\mathbf{q}_\parallel -},$$

$$\begin{aligned}\Gamma^+_n = {} & \frac{\pi I_n^2}{4N'}\sum_{\mathbf{q}_\parallel}\big(\bar{N}^n_{\mathbf{q}_\parallel - \mathbf{k}_\parallel} + 2\langle S^z\rangle_n\bar{n}^n_{\mathbf{q}_\parallel -}\big)\delta\big(E^n_{\mathbf{q}_\parallel - \mathbf{k}_\parallel} - \epsilon^n_{\mathbf{q}_\parallel -} + \epsilon^n_{\mathbf{k}_\parallel +}\big) \\ & + \Bigg[\frac{\pi I_n^2\langle S^z\rangle_n}{2N'^2}\sum_{\mathbf{q}_\parallel\mathbf{p}_\parallel}\big(\bar{N}^n_{\mathbf{p}_\parallel + \mathbf{q}_\parallel - \mathbf{k}_\parallel} + \bar{N}^n_{\mathbf{p}_\parallel}\big)\big(1 - \bar{n}^n_{\mathbf{q}_\parallel +}\big) \\ & + \frac{\pi I_n^2}{4N'^2}\sum_{\mathbf{q}_\parallel\mathbf{p}_\parallel}\bar{N}^n_{\mathbf{p}_\parallel + \mathbf{q}_\parallel - \mathbf{k}_\parallel}\big(2\langle S^z\rangle_n + \bar{N}^n_{\mathbf{p}_\parallel}\big)\Bigg] \\ & * \delta\big(E^n_{\mathbf{p}_\parallel} - E^n_{\mathbf{p}_\parallel + \mathbf{q}_\parallel - \mathbf{k}_\parallel} - \epsilon^n_{\mathbf{q}_\parallel +} + \epsilon^n_{\mathbf{k}_\parallel +}\big) \\ & + \frac{\pi}{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 -}\big(1 - \bar{n}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} - \bar{n}^n_{\mathbf{p}_\parallel + \mathbf{q}_\parallel -}\big) \\ & + \bar{n}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +}\bar{n}^n_{\mathbf{p}_\parallel + \mathbf{q}_\parallel -}\big]\delta\big(\epsilon^n_{\mathbf{p}_\parallel -} - \epsilon^n_{\mathbf{p}_\parallel + \mathbf{q}_\parallel -} - \epsilon^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} + \epsilon^n_{\mathbf{k}_\parallel +}\big) \\ & + \frac{\pi}{N'}\sum_{\mathbf{q}_\parallel}F_n^2(\mathbf{q}_\parallel)\big[\big(\bar{m}^n_{\mathbf{q}_\parallel} + 1 - \bar{n}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +}\big)\delta\big(\epsilon^n_{\mathbf{k}_\parallel +} - \epsilon^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} - \bar{\omega}^n_{\mathbf{q}_\parallel}\big) \\ & + \big(\bar{n}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} + \bar{m}^n_{\mathbf{q}_\parallel}\big)\delta\big(\epsilon^n_{\mathbf{k}_\parallel +} - \epsilon^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} + \bar{\omega}^n_{\mathbf{q}_\parallel}\big)\big],\end{aligned}$$

$v_n(\mathbf{k}_\parallel, \mathbf{q}_\parallel, \mathbf{p}_\parallel) = (v_{\mathbf{q}_\parallel} + v_{\mathbf{k}_\parallel - \mathbf{q}_\parallel - \mathbf{p}_\parallel}) - (v_{\mathbf{k}_\parallel - \mathbf{q}_\parallel} + v_{\mathbf{p}_\parallel + \mathbf{q}_\parallel})$,
$\gamma(\mathbf{k}_\parallel) = \frac{1}{2}(\cos(k_x a) + \cos(k_y a))$.

$t_s = -W_s$ and $t = -W$, where W_s and W are the conduction band width for $n = 1, N$ and for $n = 2, \ldots, N-1$, respectively. $\bar{N}_{\mathbf{q}_\parallel} = \langle S^+_{\mathbf{q}_\parallel} S^-_{\mathbf{q}_\parallel} \rangle$, $\bar{n}_{\mathbf{q}_\parallel \sigma} = \langle c^+_{\mathbf{q}_\parallel \sigma} c_{\mathbf{q}_\parallel \sigma} \rangle$ and $\bar{m}_{\mathbf{q}_\parallel} = \langle a^+_{\mathbf{q}_\parallel} a_{\mathbf{q}_\parallel} \rangle$ are the spin, electron and phonon correlation functions, respectively. They are calculated 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.

At $T = 0$ the expression for the electronic damping of the ferromagnetic semiconducting thin film simplifies to

$$\Gamma^+_n(T=0) = \frac{\pi}{N'} \sum_{\mathbf{q}_\parallel} F^2_n(\mathbf{q}_\parallel) \delta\big(\epsilon^n_{\mathbf{k}_\parallel +} - \epsilon^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} - \bar{\omega}^n_{\mathbf{q}_\parallel}\big). \tag{10}$$

Hence the electrons may be damped at zero temperature, provided that the delta function can be satisfied. In the absence of the electron–phonon interaction the electrons with $\sigma = +1$ (in the direction of the spontaneous momentum) are undamped, whereas the electrons with $\sigma = -1$ are damped.

Above T_C the magnetizations vanish. As a consequence, the expression for the electronic damping is simpler than below T_C:

$$\begin{aligned}\Gamma^+_n(T \geqslant T_C) = {} & \frac{\pi}{N'^2} \sum_{\mathbf{q}_\parallel \mathbf{p}_\parallel} v^2_n(\mathbf{k}_\parallel, \mathbf{q}_\parallel, \mathbf{p}_\parallel) \big[\bar{n}^n_{\mathbf{p}_\parallel -} \big(1 - \bar{n}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} - \bar{n}^n_{\mathbf{p}_\parallel + \mathbf{q}_\parallel -}\big) \\ & + \bar{n}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} \bar{n}^n_{\mathbf{p}_\parallel + \mathbf{q}_\parallel -}\big] \delta\big(\epsilon^n_{\mathbf{p}_\parallel -} - \epsilon^n_{\mathbf{p}_\parallel + \mathbf{q}_\parallel -} - \epsilon^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} + \epsilon^n_{\mathbf{k}_\parallel +}\big) \\ & + \frac{\pi}{N'} \sum_{\mathbf{q}_\parallel} F^2_n(\mathbf{q}_\parallel) \big[\big(\bar{m}^n_{\mathbf{q}_\parallel} + 1 - \bar{n}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +}\big) \delta\big(\epsilon^n_{\mathbf{k}_\parallel +} - \epsilon^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} - \bar{\omega}^n_{\mathbf{q}_\parallel}\big) \\ & + \big(\bar{n}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} + \bar{m}^n_{\mathbf{q}_\parallel}\big) \delta\big(\epsilon^n_{\mathbf{k}_\parallel +} - \epsilon^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} + \bar{\omega}^n_{\mathbf{q}_\parallel}\big)\big].\end{aligned} \tag{11}$$

The electronic damping of the ferromagnetic semiconducting thin film above T_C arises from the Coulomb and the electron–phonon interaction.

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 $(\mathbf{g}_n)_m = g_{mn}$, so equation (9) yields

$$\mathbf{L}(\mathbf{E})\mathbf{g}_n = \mathbf{I}. \tag{12}$$

From equation (12), $g_{nn}(E)$ is obtained as

$$g_{nn}(E) = \frac{|L_{nn}(E)|}{|L(E)|}, \tag{13}$$

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 function $g_{nn}(E)$ can be obtained by solving $|L(E)| = 0$.

The layer conduction electron magnetization can be calculated from

$$\langle s^z_n \rangle = \frac{f_{n+} - f_{n-}}{2} = \frac{1}{2N'} \sum_{\mathbf{q}_\parallel, \sigma} \sigma \langle c^+_{\mathbf{q}_\parallel \sigma} c_{\mathbf{q}_\parallel \sigma} \rangle_n; \qquad \sigma = \pm 1, \tag{14}$$

where f_{n+} and f_{n-} are the numbers of conduction electrons in the spin-up and spin-down bands of the nth layer, respectively. So, through the renormalized conduction electron energy and the conduction electron magnetization $\langle s^z_n \rangle$ we take into account the t dependence of the spin wave spectrum and our theoretical results can be applied to narrow-band as well as to wide-band ferromagnetic semiconductors. Equation (14) containing the localized spin magnetization $\langle S^z_n \rangle$, too, has to be calculated numerically. Due to the assumption of symmetrical surfaces,

there are $N/2$ equations for $N/2$ layer magnetizations which have to be solved self-consistently. Above T_C all magnetizations are zero.

The relative localized spin magnetization $M_n = \langle S_n^z \rangle$ of the nth layer is calculated from the Green's function for the magnetic excitations $G_{ij}(t) = \langle\langle S_i^+(t);\, S_j^-(0)\rangle\rangle$, obtained in [11].

4. The phonon Green function

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

$$G_{ij}(t) = \langle\langle a_i(t); a_j^+(0)\rangle\rangle. \tag{15}$$

Analogously to the case in the previous section, after a two-dimensional Fourier transformation we get for the matrix Green function the following expression:

$$\mathbf{H}(\omega)\mathbf{G}(\mathbf{k}_\parallel, \omega) = \mathbf{I}, \tag{16}$$

where

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

with

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

$$\begin{aligned} V_n = {} & \omega_{\mathbf{k}_\parallel} - R_n(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \mathbf{k}_\parallel)\langle S^z\rangle_n^2 - R_{n-1}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \mathbf{k}_\parallel)\langle S^z\rangle_{n-1}^2 - R_{n+1}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \mathbf{k}_\parallel)\langle S^z\rangle_{n+1}^2 \\ & + B_n(\mathbf{k}_\parallel, \mathbf{k}_\parallel)\langle Q_{\mathbf{k}_\parallel}\rangle_n\delta_{\mathbf{k}_\parallel 0} + B_{n-1}(\mathbf{k}_\parallel, \mathbf{k}_\parallel)\langle Q_{\mathbf{k}_\parallel}\rangle_{n-1}\delta_{\mathbf{k}_\parallel 0} \\ & + B_{n+1}(\mathbf{k}_\parallel, \mathbf{k}_\parallel)\langle Q_{\mathbf{k}_\parallel}\rangle_{n+1}\delta_{\mathbf{k}_\parallel 0}, \end{aligned}$$

$$\begin{aligned} \gamma_n = {} & \frac{4\pi\langle s^z\rangle_n^2}{N'^2}\sum_{\mathbf{q}_\parallel} F_n^2(\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(\epsilon^n_{\mathbf{q}_\parallel +} - \epsilon^n_{\mathbf{q}_\parallel - \mathbf{k}_\parallel +} + \bar{\omega}^n_{\mathbf{k}_\parallel}\big) \\ & + \frac{4\pi\langle s^z\rangle_n^2}{N'^2}\sum_{\mathbf{q}_\parallel, \mathbf{p}_\parallel}\big(R_n^2(-\mathbf{k}_\parallel, \mathbf{p}_\parallel, \mathbf{q}_\parallel)(\bar{n}^n_{\mathbf{q}_\parallel +} - \bar{n}^n_{\mathbf{p}_\parallel +})\big[(1 + \bar{m}^n_{\mathbf{k}_\parallel + \mathbf{p}_\parallel - \mathbf{q}_\parallel}) \\ & * \delta\big(\epsilon^n_{\mathbf{p}_\parallel +} - \epsilon^n_{\mathbf{q}_\parallel +} - \bar{\omega}^n_{\mathbf{k}_\parallel + \mathbf{p}_\parallel - \mathbf{q}_\parallel} + \bar{\omega}^n_{\mathbf{k}_\parallel}\big) + \bar{m}^n_{\mathbf{q}_\parallel - \mathbf{k}_\parallel - \mathbf{p}_\parallel} \\ & * \delta\big(\epsilon^n_{\mathbf{p}_\parallel +} - \epsilon^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_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]\bar{n}^n_{\mathbf{q}_\parallel +}(1 + \bar{n}^n_{\mathbf{p}_\parallel +}) \\ & * \big[\delta\big(\epsilon^n_{\mathbf{p}_\parallel +} - \epsilon^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(\epsilon^n_{\mathbf{p}_\parallel +} - \epsilon^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) \\ & + \frac{3\pi}{N'}\sum_{\mathbf{q}_\parallel}\big[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)\big] \\ & * (\bar{m}^n_{\mathbf{q}_\parallel} - \bar{m}^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{\pi}{N'}\sum_{\mathbf{q}_\parallel} A_n^2(\mathbf{q}_\parallel)\big[\big(\bar{m}^n_{\mathbf{q}_\parallel} + 1 - \bar{n}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +}\big)\delta\big(\epsilon^n_{\mathbf{k}_\parallel +} - \epsilon^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} - \bar{\omega}^n_{\mathbf{q}_\parallel}\big) \\ & + \big(\bar{n}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} + \bar{m}^n_{\mathbf{q}_\parallel}\big)\delta\big(\epsilon^n_{\mathbf{k}_\parallel +} - \epsilon^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} + \bar{\omega}^n_{\mathbf{q}_\parallel}\big)\big], \end{aligned}$$

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

At $T = 0$, where the part of the phonon damping due to the phonon–phonon interaction vanishes, we obtain

$$\gamma_n(T=0) = \frac{4\pi \langle s^z \rangle_n^2}{N'^2} \sum_{\mathbf{q}_\parallel, \mathbf{p}_\parallel} \left[R_n^2(-\mathbf{k}_\parallel, \mathbf{p}_\parallel, \mathbf{q}_\parallel) + R_n^2(-\mathbf{k}_\parallel - \mathbf{q}_\parallel + \mathbf{p}_\parallel, \mathbf{p}_\parallel, \mathbf{q}_\parallel) \right]$$
$$* \left[\delta\left(\epsilon^n_{\mathbf{p}_\parallel +} - \epsilon^n_{\mathbf{q}_\parallel +} - \bar{\omega}^n_{\mathbf{k}_\parallel + \mathbf{p}_\parallel - \mathbf{q}_\parallel} + \bar{\omega}^n_{\mathbf{k}_\parallel}\right) - \delta\left(\epsilon^n_{\mathbf{p}_\parallel +} - \epsilon^n_{\mathbf{q}_\parallel +} + \bar{\omega}^n_{\mathbf{q}_\parallel - \mathbf{k}_\parallel - \mathbf{p}_\parallel} + \bar{\omega}^n_{\mathbf{k}_\parallel}\right) \right]. \quad (17)$$

It is seen that at $T = 0$ the phonon modes of the thin film are damped due to the electron–phonon anharmonic interaction if the δ functions can be satisfied.

In the vicinity of T_C and above it, $T \geqslant T_C$, only the phonon–phonon anharmonic terms contribute to the phonon damping:

$$\gamma_n(T \geqslant T_C) = \frac{3\pi}{N'} \sum_{\mathbf{q}_\parallel} \left[B_n^2(\mathbf{q}_\parallel, -\mathbf{k}_\parallel, \mathbf{k}_\parallel - \mathbf{q}_\parallel) + B_n^2(\mathbf{q}_\parallel, \mathbf{k}_\parallel - \mathbf{q}_\parallel, -\mathbf{k}_\parallel) \right]$$
$$* (\bar{m}^n_{\mathbf{q}_\parallel} - \bar{m}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel})[\delta(\bar{\omega}^n_{\mathbf{k}_\parallel} - \bar{\omega}^n_{\mathbf{q}_\parallel} - \bar{\omega}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel}) + \delta(\bar{\omega}^n_{\mathbf{k}_\parallel} - \bar{\omega}^n_{\mathbf{q}_\parallel} + \bar{\omega}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel})]$$
$$+ \frac{\pi}{N'} \sum_{\mathbf{q}_\parallel} A_n^2(\mathbf{q}_\parallel) \left[\left(\bar{m}^n_{\mathbf{q}_\parallel} + 1 - \bar{n}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +}\right) \delta\left(\epsilon^n_{\mathbf{k}_\parallel +} - \epsilon^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} - \bar{\omega}^n_{\mathbf{q}_\parallel}\right) \right.$$
$$\left. + \left(\bar{n}^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} + + \bar{m}^n_{\mathbf{q}_\parallel}\right) \delta\left(\epsilon^n_{\mathbf{k}_\parallel +} - \epsilon^n_{\mathbf{k}_\parallel - \mathbf{q}_\parallel +} + \bar{\omega}^n_{\mathbf{q}_\parallel}\right) \right]. \quad (18)$$

We obtain solutions of the matrix equation (16) analogous to those of the previous section.

5. Numerical results and discussion

Firstly we will study the effects of the electron–electron and electron–phonon interactions on the electronic energy and damping of thin ferromagnetic semiconducting films. Thin films are of particular interest because their critical properties are more affected by surface parameters than thicker films. The temperature dependence of the renormalized electronic energy is calculated numerically for a simple cubic (sc) thin ferromagnetic semiconducting film with parameters for $CdCr_2Se_4$ [24]: $J_s = 0.2J$, $D_s = D = 0.01$ 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, $B = -2.54$ cm^{-1}, $F = 23$ cm^{-1}, $R = -18$ cm^{-1}, $A = 6.61$ cm^{-1}, $S = 3/2$, $W = 0.1$ eV, $U \equiv 0.5v(\mathbf{k}) = 0.3$ 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 2–3 greater than in the bulk [25, 26]. Therefore we have chosen greater surface anharmonic constants compared to the bulk ones. It will be shown that the enhanced surface anharmonicity leads to a decrease in energy and increase in width of a surface phonon.

With increasing Coulomb interaction constant the electronic energy increases, too. This is valid for the spin-down ($\sigma = -1$) band energy for all temperatures and for all bandwidth values W. For narrow bands $W \ll U$ (for example $W = 0.1$ eV) the Coulomb interaction constant U does not influence the spin-up ($\sigma = +1$) band energy at low temperatures. For $W \geqslant U$ (for example $W = 1$ eV), at low temperatures, U increases the spin-up band energy for $|W - 0.5IS| \geqslant U$ and decreases it for $|W - 0.5IS| < U$. Above T_C the electronic energy increases with U.

The electronic spectrum is renormalized at $T = 0$ due to the s–d and electron–phonon interaction. In the absence of the electron–phonon interaction only the energy of the spin-down band is renormalized at $T = 0$ due to the s–d interaction. In figure 1 the temperature dependence of the electronic spectrum is presented for a thin ($N = 8$ layers) film. The surface mode is clearly distinguished (curve 1, $n = 1$) from the bulk modes (curve 2, 3 and 4, $n = 2$, 3 and 4). With increase of the surface spin–spin J_s or s–d I_s interaction constants, the difference

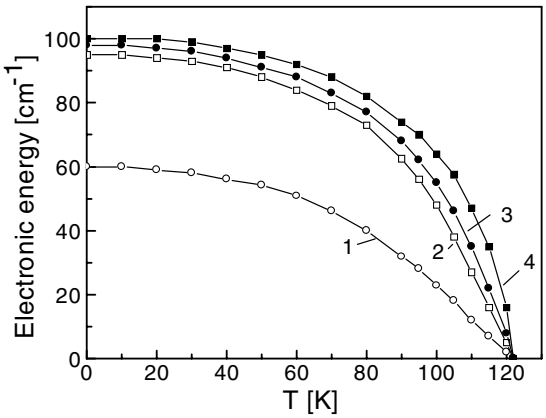

Figure 1. The temperature dependence of the electronic energy ϵ for a ferromagnetic semiconducting thin film for $N = 8$ and different layers: (1) $n = 1$, (2) 2, (3) 3, (4) 4.

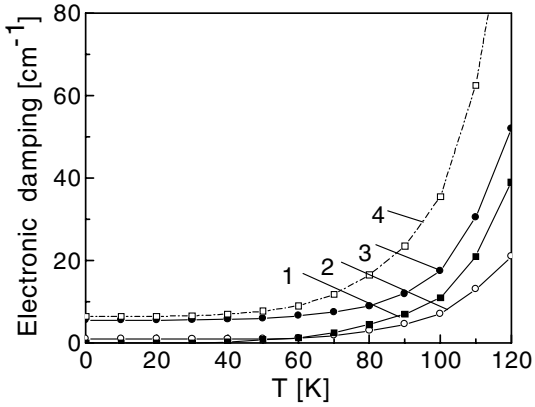

Figure 2. The temperature dependence of the electronic damping $\Gamma^{+}_{\rm el}$ for a ferromagnetic semiconducting thin film for $N = 8$: (1) $\Gamma^{+}_{\rm el-el}$, (2) $\Gamma^{+}_{\rm s-d}$, (3) $\Gamma^{+}_{\rm el-ph}$, (4) $\Gamma^{+}_{\rm el}$.

between the surface and the bulk modes is enhanced. The calculations show that the electron–phonon interaction lowers the surface energy and the energy of the thin film, and cannot be neglected. The electronic energy of the film with $N > 30$ layers coincides with that for the bulk.

The electronic damping $\Gamma^{\sigma}_{\rm el}$ of the thin film is larger compared to the bulk one. At low temperatures the damping is very small and increases very strongly with increasing $T \to T_{\rm C}$ (figure 2). The scattering terms give more important contributions to the damping than the decay terms. The former terms are of higher order in $1/z$ than the latter terms, but they are usually omitted in an expansion with respect to powers of $1/z$ (z is the number of the nearest neighbours). Here we have no $1/z$ expansion and so we have taken into account all summation terms. $\Gamma^{\sigma}_{\rm el}$ contains the following terms due to the different interactions:

$$\Gamma^{\sigma}_{\rm el} = \Gamma^{\sigma}_{\rm s-d} + \Gamma^{\sigma}_{\rm el-el} + \Gamma^{\sigma}_{\rm el-ph}. \tag{19}$$

The delta functions in $\Gamma^{\sigma}_{\rm el-ph}$ can be satisfied only for small $\mathbf{k}$, ($\mathbf{k} \approx 0$). For all temperatures this term is very large compared with the other two terms, which is in agreement with the experimental data of Rehbein *et al* [18], where the observed increase of the linewidth with

temperature is attributed to enhanced electron–phonon scattering. At low temperatures the contribution in the electronic damping caused by the electron–electron interaction is very small compared with the term due to the s–d interaction. Therefore at low temperatures we obtain

$$\Gamma^{\sigma}_{\mathrm{el-el}} \ll \Gamma^{\sigma}_{\mathrm{s-d}} \ll \Gamma^{\sigma}_{\mathrm{el-ph}}, \qquad T \ll T_{\mathrm{C}}. \tag{20}$$

For $T \approx 0.5T_{\mathrm{C}}$ we have $\Gamma^{\sigma}_{\mathrm{el-el}} \approx \Gamma^{\sigma}_{\mathrm{s-d}}$, whereas at higher temperatures ($T > 0.5T_{\mathrm{C}}$) the part from the electron–electron interaction predominates over this due to the s–d interaction. So we can write

$$\Gamma^{\sigma}_{\mathrm{s-d}} \ll \Gamma^{\sigma}_{\mathrm{el-el}} \ll \Gamma^{\sigma}_{\mathrm{el-ph}}, \qquad T > 0.5T_{\mathrm{C}}. \tag{21}$$

In the vicinity of T_{C} for $W \gg U$ we have $\Gamma^{\sigma}_{\mathrm{s-d}} \gg \Gamma^{\sigma}_{\mathrm{el-el}}$, whereas for $W \ll U$ we have $\Gamma^{\sigma}_{\mathrm{s-d}} \ll \Gamma^{\sigma}_{\mathrm{el-el}}$. For $W \approx U$ we have $\Gamma^{\sigma}_{\mathrm{s-d}} \approx \Gamma^{\sigma}_{\mathrm{el-el}}$. For constant bandwidth W with increasing electron–electron interaction U the electronic damping increases, too. The temperature dependence of the lifetime broadening is mainly given by the temperature dependence of the electron–phonon scattering rate. The electron–electron scattering shows weaker temperature dependence, in agreement with the theoretical results of Rehbein *et al* [18]. The theoretical investigations of Hellsing *et al* [15] also show that the damping due to the electron–phonon coupling is greater than the damping caused by the electron–electron interaction. But unlike us, they obtain a temperature independent $\Gamma_{\mathrm{el-el}}$.

Now we will study the effects of the electron–phonon and phonon–phonon interactions on the phonon frequency and damping of thin ferromagnetic semiconducting films. The phonon energy for the phonon mode $\omega_0 = 100\ \mathrm{cm}^{-1}$ (D mode) and the damping were calculated numerically using the same parameters as for the electronic spectrum. Some interesting features are observed from the results obtained. The surface phonon energy $\bar{\omega}_{\mathrm{s}}$ is much smaller than the phonon energy of the inner layer $\bar{\omega}_{N/2}$; the surface phonon decrease is four times greater than that observed in the bulk. This is due to the lower coordination number of the surface phonons and to the electron–phonon interaction. With increasing electron–phonon interaction constant the surface phonon energy decreases, in agreement with the experimental data of Watanabe *et al* [20]. The surface damping γ_{s} is much larger compared to the damping of the inner layer $\gamma_{N/2}$. 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 electron–phonon interaction.

The temperature dependence of the renormalized phonon energy $\bar{\omega}$ is plotted in figure 3 for a sc ferromagnetic semiconducting film for different thicknesses of the film ($N = 6$ and 8 layers) with and without electron–phonon interaction. It can be seen that the electron–phonon interaction reduces the phonon energy and must be taken 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 $\bar{\omega}_{\mathrm{TF}} < \bar{\omega}_{\mathrm{B}}$, i.e. the phonon frequency of the thin film shifts to lower energy due to the existence of a surface mode and due to electron–phonon coupling. This is in agreement with the experimental data of Chzang *et al* [19]. The phonon energy of the film with $N > 30$ layers coincides with that for the bulk.

The phonon damping $\gamma_{\mathrm{ph}} = \gamma_{\mathrm{ph-ph}} + \gamma_{\mathrm{el-ph}}$ is plotted in figure 4 as a function of temperature for various film thickness ($N = 8, 16$ and 30 layers). The main signature of the electron–phonon contribution to the lifetime broadening is the temperature dependence. The electron–phonon interaction enhances the phonon damping of the thin film. The damping increases near T_{c}, reaches a maximum, and then remains nearly constant. At $T = 0$ the phonon modes are damped due to the electron–phonon interaction. Only the electron–phonon anharmonic interaction terms contribute to γ_{ph} at $T = 0$. In the vicinity of T_{c} and above it, where the magnetizations $\langle S^z \rangle = 0$ and $\langle s^z \rangle = 0$, only the phonon–phonon anharmonic terms contribute

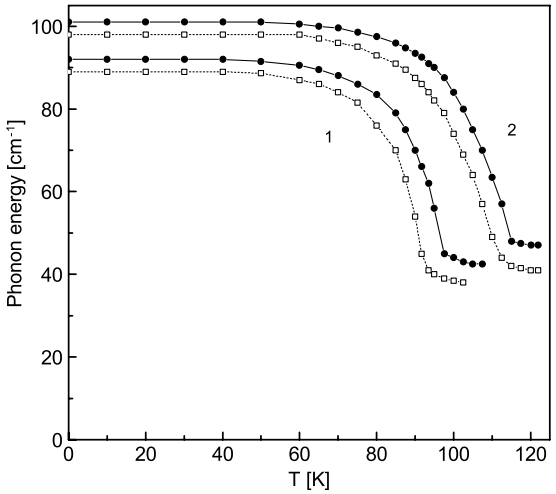

Figure 3. The temperature dependence of the phonon energy $\bar{\omega}$ for a ferromagnetic semiconducting thin film for different film thicknesses: (1) $N = 6$, (2) 8, dashed curve—with and full curve—without electron–phonon interaction.

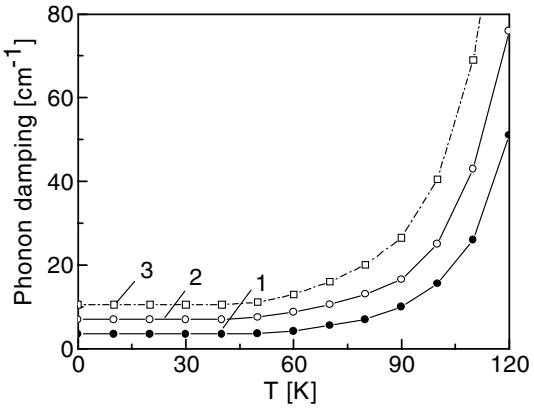

Figure 4. The temperature dependence of the phonon damping $\gamma_{\rm ph}$ for a ferromagnetic semiconducting thin film for different film thicknesses: (1) $N = 30$, (2) 16, (3) 8.

to the phonon damping. It can be seen that there are several differences between the thin film and the bulk behaviour. Thinner films have larger damping. For $N < 30$ layers we have $\gamma_{\rm TF} > \gamma_{\rm B}$; i.e. the damping is larger for thin films than that for the bulk, in agreement with the experimental data obtained for Fe thin films by Roehlsberger *et al* [4]. 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. Baddorf and Plumer [21] have shown experimentally that the anharmonicity at the surface is 4–5 times greater than that in the bulk.

6. Conclusions

Beyond the random phase approximation we get the renormalized electronic spectrum for a sc ferromagnetic semiconducting thin film. The electron–phonon coupling plays an important

role. It decreases the surface electronic energy of the film. The electronic damping is obtained and calculated numerically. At low temperatures we obtain $\Gamma^{\sigma}_{\mathrm{el-el}} \ll \Gamma^{\sigma}_{\mathrm{s-d}}$, whereas at higher temperatures we have $\Gamma^{\sigma}_{\mathrm{s-d}} \ll \Gamma^{\sigma}_{\mathrm{el-el}}$. The term $\Gamma^{\sigma}_{\mathrm{el-ph}}$ is for every temperature very large compared to the other two terms in the electronic damping, so it must be taken into account if we want to explain the origin of the lifetime broadening of electronic states at surfaces and in thin films. The damping in thin films is greater compared to the bulk case due to surface effects and electron–phonon interactions.

We have shown for the first time the importance and the influence of the electron–phonon interaction on the phonon spectrum of thin ferromagnetic semiconducting films. The phonon frequencies are smaller, whereas the damping effects are larger, in comparison to the bulk. The phonon modes display a non-linear temperature dependence due to the fourth-order anharmonic phonon–phonon interaction and the third-order anharmonic electron–phonon interaction. At low temperatures the electron–phonon anharmonic terms play an important role in the phonon damping, whereas at $T \geqslant T_{\mathrm{c}}$ the anharmonic phonon–phonon interaction is important. The observed increase of the linewidth with temperature is attributed to enhanced electron–phonon scattering.

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