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Anharmonic effects in ferromagnetic semiconductors

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Abstract. A Green function technique is used to study the anharmonic spin–phonon and phonon–phonon interaction effects on optical phonon modes and spin-wave excitations in ferromagnetic semiconductors. The cubic spinels have been investigated because the magnetostriction of these compounds is small and the direct contribution of spin ordering to the phonon modes can be clearly observed. The phonon and spin-wave energy and damping are evaluated for the first time beyond the random-phase approximation. The temperature dependence of these quantities is discussed and is found to be in good agreement with the experimental data.

1. Introduction

In recent years, the effects of spin ordering on phonon properties have been investigated for many magnetic crystals, such as the Cr spinels, EuX (X = O, S, Te or Se) and fluorite-type compounds [1, 2]. The phonon frequency and the phonon damping have been determined from the spectra. The effects of spin ordering on the phonon modes in ACr_2X_4 were directly observed from experiments. In the spinels these values showed anomalous shifts characterizing the spin ordering effect [2]. For infrared-active phonons in cubic antiferromagnetic $KNiF_3$, Sintani *et al* [3] observed a small but rapid frequency shift at the Curie temperature T_C. They assigned this to the effect of spin ordering and evaluated the change in the force constant modulated by the spin ordering. For several cubic spinels, Wakamura and Arai [4] observed the temperature dependence of ω_{l0} and ω_{t0} for all the infrared-active modes. These modes exhibit a rapid frequency shift at T_C and an appreciable shift to a temperature T_0, which is higher than T_C [4]. We made an attempt to explain these anomalies theoretically, on the basis of the anharmonic phonon–phonon interaction. It was established that above T_C, i.e. in the higher-temperature region, there exists good agreement with the experimental data for magnetic crystals and in the whole temperature region for non-magnetic crystals. However, below T_C there is a large deviation in the theoretical curves from the experimental curves, which could be attributed to the spin ordering effect. Only the interaction between the spin and phonon subsystems may explain these phenomena.

There are only a few papers which consider magnon damping in ferromagnetic semiconductors, taking into account the spin–phonon interaction. In [5–7] the calculation generally employs the Holstein–Primakoff transformation of the spin operators and is valid at low temperatures $T \ll T_C$, where this type of interaction describes two-magnon–one-phonon processes. The Green function methods and the random-phase approximation (RPA) were used in [8, 9] for temperatures below T_C. Wesselinowa and Apostolov [10] have studied the effects of the spin–phonon interaction on the spin-wave energy and transverse damping.

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Spin-wave damping was observed experimentally for $CdCr_2Se_4$ and $CdCr_2S_4$ by Anisimov and Green [11] and for EuO by Gurevich and Anisimov [12]. They found that the spin-wave damping increases strongly when the temperature increases, and that the magnon damping is nearly independent of the wavevector $\boldsymbol{k}$ in a wide region ($|\boldsymbol{k}| = 0$–2×10^6 cm^{-1}) at low temperatures.

The aim of the present paper is to extend our previous work [10] and to study theoretically the influence of the anharmonic spin–phonon interaction on the phonon and spin-wave spectrum in cubic spinels.

2. Model and method

The full Hamiltonian for the anharmonic magnetic crystal may be written as

$$H = H_d + H_s + H_{sd} + H_{ph} + H_{sp}. \tag{1}$$

H_d is the Heisenberg Hamiltonian for the ferromagnetically ordered electrons:

$$H_d = -\tfrac{1}{2} \sum_q J_q (S_q^z S_{-q}^z + S_q^- S_q^+) - g\mu_B \mathbb{H} \sqrt{N} S_0^z. \tag{2}$$

H_s represents the usual Hamiltonian of the conduction band electrons:

$$H_s = \sum_{q,\sigma} (\varepsilon_{q\sigma} - \mu) c_{q\sigma}^+ c_{q\sigma} \tag{3}$$

where $\sigma = \pm 1$, $c_{q\sigma}$ and $c_{q\sigma}^+$ are the Fermi operators of annihilation and creation, respectively, in the state $q\sigma$, μ is the chemical potential and $\varepsilon_{q\sigma}$ are the Bloch energies.

H_{sd} is the s–d interaction term which couples the two subsystems (2) and (3) by an intratomic exchange interaction:

$$H_{sd} = -\frac{I}{2N} \sum_{q,p} [S_{q-p}^+ c_{p-}^+ c_{q+} + S_{q-p}^- c_{p+}^+ c_{q-} + S_{q-p}^z (c_{p+}^+ c_{q+} - c_{p-}^+ c_{q-})] \tag{4}$$

where I is the interaction constant.

H_{ph} contains the lattice vibrations including third- and fourth-order anharmonic phonon–phonon interactions:

$$\begin{aligned} H_{ph} = {} & \frac{1}{2!} \sum_q (P_q P_{-q} + \omega_q^2 Q_q Q_{-q}) + \frac{1}{3!} \sum_{q,q_1} B(q, q_1) Q_q Q_{-q_1} Q_{q_1-q} \\ & + \frac{1}{4!} \sum_{q,q_1,q_2} A(q, q_1, q_2) Q_{q_1} Q_{q_2} Q_{-q-q_2} Q_{-q_1+q} \end{aligned} \tag{5}$$

where Q_q, P_q and ω_q are the normal coordinate, momentum and frequency, respectively, of the lattice mode with a wavevector $\mathbf{q}$. The vibrational normal coordinate Q_q and the momentum P_q can be expressed in terms of phonon creation and annihilation operators:

$$Q_q = (2\omega_q)^{-1/2} (a_q + a_{-q}^+) \qquad P_q = \mathrm{i}(\omega_q/2)^{1/2} (a_q^+ + a_{-q}) \tag{6}$$

where $[a_q; a_p^+]_- = \delta_{qp}$.

H_{sp} describes the interaction of the spin with the phonons:

$$\begin{aligned} H_{sp} = {} & -\tfrac{1}{2} \sum_{q,p} F(p, q) Q_{p-q} (S_q^z S_{-p}^z + S_q^- S_p^+) \\ & -\tfrac{1}{4} \sum_{q,p,\nu} R(\nu, p, q) Q_\nu Q_{p-q-\nu} (S_q^z S_{-p}^z + S_q^- S_p^+) \\ & -\tfrac{1}{12} \sum_{q,p,\nu,\mu} T(\mu, \nu, p, q) Q_\mu Q_\nu Q_{p-q-\nu-\mu} (S_q^z S_{-p}^z + S_q^- S_p^+) \end{aligned} \tag{7}$$

where

$$F(p,q)=\frac{1}{\sqrt{N}}\sum_h\frac{1}{|h|}\frac{(\boldsymbol{e}_{p-q}\cdot\boldsymbol{h})}{(2\omega_{p-q})^{1/2}}J'(h)(\exp(\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{h})+\exp(\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{h}))$$

$$R(\nu,p,q)=\frac{1}{N}\sum_h\left[\left(J''(h)-\frac{J'(h)}{|h|}\right)(\boldsymbol{e}_\nu\cdot\boldsymbol{h})(\boldsymbol{e}_{p-q-\nu}\cdot\boldsymbol{h})+\frac{J'(h)}{|h|}(\boldsymbol{e}_\nu\cdot\boldsymbol{e}_{p-q-\nu})\right]$$
$$\times(1-\exp(\mathrm{i}\boldsymbol{\nu}\cdot\boldsymbol{h}))(\exp(\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{h})+\exp(\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{h}))(4\omega_\nu\omega_{p-q-\nu})^{-1/2}$$

$$T(\mu,\nu,p,q)=\frac{1}{N^{3/2}}\sum_h\left\{\left[J'''(h)-\left(\frac{2}{|h|}+\frac{1}{|h|^2}\right)J''(h)+\frac{4J'(h)}{|h|^3}\right](\boldsymbol{e}_\nu\cdot\boldsymbol{h})(\boldsymbol{e}_\mu\cdot\boldsymbol{h})\right.$$
$$\left.\times(\boldsymbol{e}_{p-q-\nu-\mu}\cdot\boldsymbol{h})+\left(\frac{J''(h)}{|h|^2}-\frac{J'(h)}{|h|^3}\right)(\boldsymbol{e}_\nu\cdot\boldsymbol{h})(\boldsymbol{e}_\mu\cdot\boldsymbol{e}_{p-q-\nu-\mu})\right\}$$
$$\times(1-\exp(\mathrm{i}\boldsymbol{\nu}\cdot\boldsymbol{h}))(1-\exp(\mathrm{i}\boldsymbol{\mu}\cdot\boldsymbol{h}))(\exp(\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{h})+\exp(\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{h}))$$
$$\times(8\omega_\nu\omega_\mu\omega_{p-q-\nu-\mu})^{-1/2}.$$

The summations extend over the vectors $\boldsymbol{h}=\boldsymbol{r}_i-\boldsymbol{r}_j$ connecting all possible pairs of spin sites in the crystal and $\boldsymbol{e}_q$ is the polarization of the phonon with wavevector $\boldsymbol{q}$.

The retarded Green function to be calculated is defined in matrix form as

$$G_k=-\mathrm{i}\theta(t)\langle[B_k(t);B_k^+]_-\rangle. \tag{8}$$

The operator B_k stands symbolically for the set S_k^+; $\sum_p c^+_{p+k+}c_{p-}$; a_k; a^+_{-k}. For the approximate calculation of the Green function (8) we use a method proposed by Tserkovnikov [13], which is appropriate for spin problems.

After a formal integration of the equation of motion for (8), one obtains

$$G_k(t)=-\mathrm{i}\theta(t)\langle[B_k;B_k^+]_-\rangle\exp(-\mathrm{i}\Omega_k(t)t) \tag{9}$$

where

$$\Omega_k(t)=\Omega_k-\frac{\mathrm{i}}{t}\int_0^t\mathrm{d}t'\,t'\left(\frac{\langle[j_k(t);j_k^+(t')]_-\rangle}{\langle[B_k(t);B_k^+(t')]_-\rangle}-\frac{\langle[j_k(t);B_k^+(t')]_-\rangle\langle[B_k(t);j_k^+(t')]_-\rangle}{\langle[B_k(t);B_k^+(t')]_-\rangle^2}\right) \tag{10}$$

with the notation $j_k(t)=\langle[B_k;H_{int}]_-\rangle$. The time-independent term

$$\Omega_k=\frac{\langle[[B_k;H]_-;B_k^+]_-\rangle}{\langle[B_k;B_k^+]_-\rangle} \tag{11}$$

is a 4×4 matrix. If we neglect the time-dependent term at $\Omega_k(t)$ for the Green function, we obtain

$$\begin{aligned}G_k(t)&=-\mathrm{i}\theta(t)\langle[B_k;B_k^+\rangle\exp(-\mathrm{i}\Omega_k t)\\&=-\mathrm{i}\theta(t)\langle[B_k;B_k^+]\rangle U_k\exp(-\mathrm{i}W_k t)U_k^{-1}\end{aligned} \tag{12}$$

where the matrix U_k consists of columns, which are the eigenvectors of Ω_k and $W_k=U_k^{-1}\Omega_k U_k$ is a diagonal matrix with components the roots of the characteristic equation for Ω_k:

$$\det|\Omega_k-WI|=0 \tag{13}$$

where I is the unit matrix. The roots of (13) determine the energy of the coupled mode in the generalized Hartree–Fock approximation. The time-dependent term includes damping effects.

3. The phonon spectrum

3.1. The phonon energy

For the optical phonon energy we obtain the following expression:

$$\bar{\omega}_k = \pm[(\omega_k^{11})^2 + (\omega_k^{12})^2]^{1/2} \tag{14}$$

where

$$\begin{aligned}\omega_k^{11} = \omega_k &- \sum_q R(k,q,q)(\langle S_q^z S_{-q}^z\rangle + \langle S_q^- S_q^+\rangle) \\ &- \sum_{q,p} T(k,-k,p,q)\langle Q_{p-q}\rangle(\langle S_q^z S_{-p}^z\rangle + \langle S_q^- S_p^+\rangle) + B(k,-k,0)\langle Q_0\rangle \\ &+ \frac{1}{2N}\sum_q \{A(k,-k,q,-q) + A(q,-k,-q,k)\}(2\bar{N}_q + 1)\end{aligned} \tag{15}$$

$$\begin{aligned}\omega_k^{12} = &\sum_q R(k,q,q)(\langle S_q^z S_{-q}^z\rangle + \langle S_q^- S_q^+\rangle) \\ &+ \sum_{q,p} T(k,-k,p,q)\langle Q_{p-q}\rangle(\langle S_q^z S_{-p}^z\rangle + \langle S_q^- S_p^+\rangle) - B(k,-k,0)\langle Q_0\rangle \\ &- \frac{1}{2N}\sum_q \{A(k,-k,q,-q) + A(q,-k,-q,k)\}(2\bar{N}_q + 1)\end{aligned} \tag{16}$$

where

$$\bar{N}_q = \langle a_q^+ a_q\rangle = 1/[\exp(\bar{\omega}_q/T) - 1] \tag{17}$$

and

$$\begin{aligned}\langle Q_{p-q}\rangle = &\Bigg(\sum_{q_1} F(p-q-q_1,q_1)(\langle S_{q_1}^z S_{q+q_1-p}^z\rangle + \langle S_{q_1}^- S_{p-q_1-q}^+\rangle) \\ &+ \frac{1}{N}\sum_{q_1,p_1} T(q-p,\nu,p_1,q_1)(1 + 2\langle a_\nu a_{p_1+q_1+\nu}^+\rangle)(\langle S_{q_1}^z S_{-p_1}^z\rangle + \langle S_{q_1}^- S_{p_1}^+\rangle) \\ &- \frac{1}{N}\sum_{q_1} B(q_1,p-q-q_1,p-q)(1 + 2\langle a_{q+q_1-p}^+ a_{q_1}\rangle)\Bigg) \\ &\times\Bigg(\omega_{p-q} - \sum_{q_1} R(q-p,q_1,q_1)(\langle S_{q_1}^z S_{-q_1}^z\rangle + \langle S_{q_1}^- S_{q_1}^+\rangle) \\ &+ \frac{1}{N}\sum_{q_1}\{A(p-q,q-p,q_1,-q_1) \\ &+ A(q_1,-q_1,p-q,q-p)\}(2\bar{N}_{q_1} + 1)\Bigg)^{-1}.\end{aligned} \tag{18}$$

If we neglect the transverse correlation function $\langle S_q^- S_q^+\rangle$ and decouple the longitudinal correlation function $\langle S_q^z S_{-q}^z\rangle \to \langle S_0^z\rangle^2\delta_{q0}$ (RPA) for $\bar{\omega}_k$, we obtain

$$\bar{\omega}_k^2 = (\omega_k)^2 - 2\omega_k\left(R_k\langle S^z\rangle^2 + T_k\langle S^z\rangle^2\langle Q_k\rangle\delta_{k0} - \frac{1}{2N}\sum_q A_{qk}(2\bar{N}_q + 1) - B_k\langle Q_k\rangle\delta_{k0}\right) \tag{19}$$

where

$$\langle Q_k\rangle = \left(F_k\langle S^z\rangle^2 + \frac{1}{N}\sum_q T_{kq}(2\bar{N}_q + 1)\langle S^z\rangle^2 - \frac{1}{N}\sum_q B_{kq}(2\bar{N}_q + 1)\right)$$

$$\times\left(\omega_k - R_k\langle S^z\rangle^2 + \frac{1}{N}\sum_q A_{kq}(2\bar{N}_q+1)\right)^{-1}. \tag{20}$$

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 numerically the temperature dependence of the phonon energy for the phonon modes $\omega_0 = 100$ cm^{-1} (D mode) and $\omega_0 = 381$ cm^{-1} (A mode) [4], for $CdCr_2Se_4$ using the following model parameters: $I = 0.5$ eV, $J_0 = 0.0001$ eV, $W = 1.0$ eV, $S = 1.5$, $B = -2.54$ cm^{-1}, $A = 6.61$ cm^{-1}, $F = 23$ cm^{-1}, $T = 1.4$ cm^{-1} and different values of R. These modes display a non-linear temperature dependence (figures 1–3). Their temperature dependence is in very good agreement with the experimental data [4].

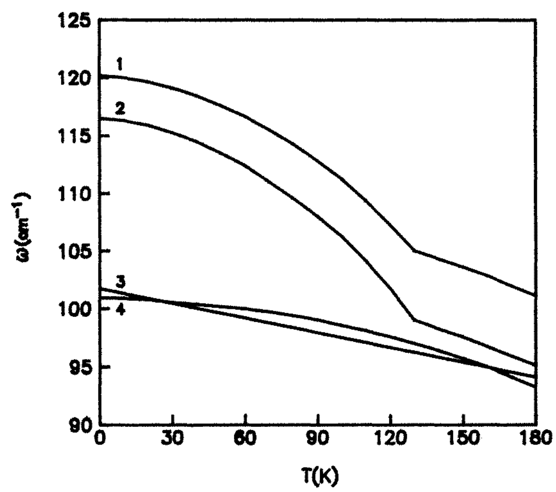

Figure 1. Temperature dependence of the phonon D mode with $\omega_0 = 100$ cm^{-1} with the following model parameters: curve 1, $B = -2.54$ cm^{-1}, $A = 6.61$ cm^{-1}, $F = 23$ cm^{-1}, $R = -18$ cm^{-1}, $T = 1.4$ cm^{-1}; curve 2, $B = -2.54$ cm^{-1}, $A = 6.61$ cm^{-1}, $F = 23$ cm^{-1}, $R = T = 0$; curve 3, $B = -2.54$ cm^{-1}, $A = F = R = T = 0$; curve 4, $B = -2.54$ cm^{-1}, $A = 6.61$ cm^{-1}, $F = R = T = 0$.

From figure 1 we may conclude that firstly, if we take into account only the third-order phonon–phonon interaction, we obtain a linear temperature dependence (curve 3) and secondly using only the anharmonic phonon–phonon interaction (curve 4) could not explain the temperature dependence below T_C (see the introduction).

Figures 2 and 3 exhibit the different behaviours of the D and A modes, respectively. This frequency shift in spinels, below T_C, can be explained only if we assume a spin-dependent force constant given by the first, second and third derivatives of the magnetic exchange interaction J_{ij} between the ith and jth magnetic ions with respect to the phonon displacements u_i, u_j. This displacement is interpreted by taking the nearest-neighbour ferromagnetic exchange integral J_{ij} and the next-nearest-neighbour antiferromagnetic exchange integral K_{ik}. J_{ij} and K_{ik} represent the integrals through the linkage of Cr–X–Cr and Cr–X–A–X–Cr, as shown by the double lines in figure 4. The magnetic properties were changed predominantly by the second-neighbour exchange interaction. The squared derivatives of J_{ij} and K_{ik} with respect to the phonon displacement have opposite signs and, if we denote them R_1 and R_2 accordingly, then the additional shift $\Delta\omega$ of the phonon

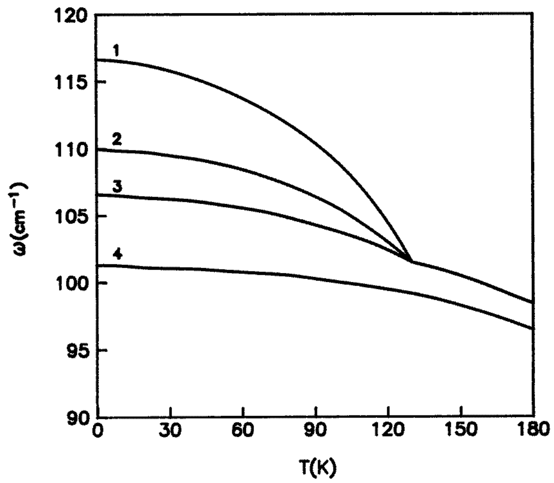

Figure 2. Temperature dependence of the phonon D mode with $\omega_0 = 100$ cm^{-1} for $B = -2.54$ cm^{-1}, $A = 6.61$ cm^{-1}, $F = 23$ cm^{-1}, $T = 1.4$ cm^{-1} and for different values of R: curve 1, $R = -15$ cm^{-1}; curve 2, $R = -12$ cm^{-1}; curve 3, $R = -7$ cm^{-1}; curve 4, $B = -2.54$ cm^{-1}, $A = 6.61$ cm^{-1}, $F = R = 0$.

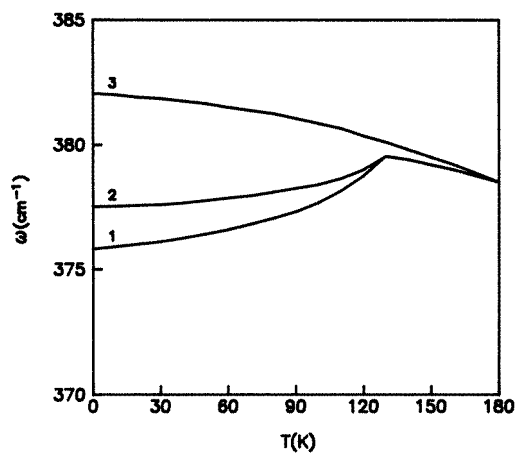

Figure 3. Temperature dependence of the phonon A mode with $\omega_0 = 381$ cm^{-1} for $B = -2.54$ cm^{-1}, $A = 6.61$ cm^{-1}, $F = 23$ cm^{-1}, $T = 1.4$ cm^{-1} and for different values of R: curve 1, $R = 3$ cm^{-1}; curve 2, $R = 1$ cm^{-1}; curve 3, $F = R = T = 0$.

frequency may be written as [1]

$$\Delta\omega = [R_1\langle S_1 S_2\rangle - R_2\langle S_1 S_3\rangle]/\langle S_0^z\rangle^2. \tag{21}$$

In our model the force constant R is equal to $R_1 - R_2$.

From figure 2 we see that the D mode exhibits a relatively large shift below T_C, which increases with increasing $|R|$. This can be explained by the large magnitude of R_2 in comparison with that of R_1, which is predicted by the large pressure dependence of K_{ik}, estimated from the Curie temperature [14, 15]. Then $R_1 - R_2 < 0$, i.e. $R < 0$.

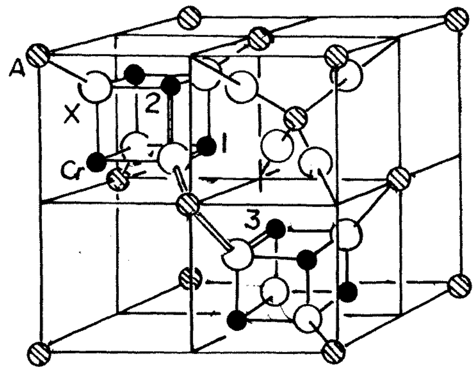

Figure 4. The unit cell of the spinel structure. The structure can be described using two types of cubic octan. The shaded and full circles indicate the A and Cr atoms, respectively. The open circles are the X atoms. The linkages of ferromagnetic Cr–X–Cr and antiferromagnetic Cr–X–A–X–Cr are depicted between Cr(1) and Cr(2) atoms and between Cr(1) and Cr(3) atoms, respectively, by double lines.

Figure 3 displays the opposite frequency shift of the A mode. This has been explained by combining the dominant contribution of the Cr ion to the A mode and the linkage of J_{ij}. In this case, R_1 has a large value. Then $R_1 - R_2$ may become positive, i.e. $R > 0$.

The calculations demonstrate that, if we want to obtain a correct temperature dependence of the phonon modes in ferromagnetic semiconductors, we must not neglect the effects of 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.

3.2. Phonon damping

In the calculations for the integral term in equation (10) which includes damping effects, we use the approximate dynamics

$$S_k(t) \sim S_k \exp(-\mathrm{i}E_k t) \qquad a_k(t) \sim a_k \exp(-\mathrm{i}\bar{\omega}_k t). \tag{22}$$

Then we obtain the following expression for phonon damping:

$$\gamma^{ph}(k) = \gamma_{sp}(k) + \gamma_{ph-ph}(k). \tag{23}$$

$\gamma_{sp}(k)$ is the damping part which comes from the spin–phonon interaction:

$$\begin{aligned}
\gamma_{sp}(k) = {} & \frac{4\pi\langle S^z\rangle^2}{N}\sum_q F^2(q, q-k)(\bar{n}_q - \bar{n}_{q-k})\delta(E_{q-k} - E_q - \bar{\omega}_k) \\
& + \frac{4\pi\langle S^z\rangle^2}{N^2}\sum_{q,p}\{R^2(-k, p, q)(\bar{n}_q - \bar{n}_p)[(1 + \bar{N}_{k+p-q}) \\
& \times\delta(E_p - E_q - \bar{\omega}_{k+p-q} + \bar{\omega}_k) + \bar{N}_{q-k-p}\delta(E_p - E_q + \bar{\omega}_{q-k-p} + \bar{\omega}_k)] \\
& + [R^2(-k, p, q) + R^2(k-q+p, p, q)]\bar{n}_q(1 + \bar{n}_p) \\
& \times[\delta(E_p - E_q - \bar{\omega}_{k+p-q} + \bar{\omega}_k) - \delta(E_p - E_q + \bar{\omega}_{q-k-p} + \bar{\omega}_k)] \\
& + \frac{\pi}{N^2}\sum_{q,p}[R^2(-k, p, q) + R^2(k-q+p, p, q)]\langle S^z_p S^z_{-p}\rangle\langle S^z_q S^z_{-q}\rangle \\
& \times[\delta(E_p - E_q - \bar{\omega}_{k+p-q} + \bar{\omega}_k) - \delta(E_p - E_q + \bar{\omega}_{q-k-p} + \bar{\omega}_k)]
\end{aligned}$$

$$
\begin{aligned}
&+\frac{4\pi\langle S^z\rangle^2}{N^3}\sum_{q,p,\nu}\{[T^2(-k,\nu,p,q)+T^2(k+p-q-\nu,\nu,p,q)]\\
&\times[\bar n_q(1+\bar n_p)(1+\bar N_\nu+\bar N_{k+p-q-\nu})+(\bar n_q-\bar n_p)\bar N_\nu\bar N_{k+p-q-\nu}]\\
&+[T(-k,\nu,p,q)T(-k,k+p-q-\nu,p,q)\\
&+T(k+p-q-\nu,\nu,p,q)T(k+p-q-\nu,k+p-q-\nu,p,q)]\\
&\times[\bar n_q(1+\bar n_p)(1+\bar N_{k+p-q-\nu})+\bar n_p(1+\bar n_q)\bar N_{k+p-q-\nu}]\}\\
&\times\delta(E_p-E_q-\bar\omega_\nu-\bar\omega_{k+p-q-\nu}+\bar\omega_k)\\
&+\frac{4\pi\langle S^z\rangle^2}{N^3}\sum_{q,p,\nu}[T^2(-k,\nu,p,q)+T^2(k+p-q-\nu,\nu,p,q)]\\
&\times\{[\bar n_q(1+\bar n_p)\bar N_{q+\nu-p-k}-\bar n_p(1+\bar n_q)\bar N_\nu+(\bar n_q-\bar n_p)\bar N_\nu\bar N_{q+\nu-p-k}]\\
&\times\delta(E_p-E_q-\bar\omega_\nu+\bar\omega_{q+\nu-p-k}+\bar\omega_k)+[\bar n_q(1+\bar n_p)\bar N_\nu-\bar n_p(1+\bar n_q)\bar N_{p+k+\nu-q}\\
&+(\bar n_q-\bar n_p)\bar N_\nu\bar N_{\nu+p+k-q}]\delta(E_p-E_q+\bar\omega_\nu-\bar\omega_{\nu+p+k-q}+\bar\omega_k)\}\\
&+\frac{4\pi\langle S^z\rangle^2}{N^3}\sum_{q,p,\nu}\{[T^2(-k,\nu,p,q)+T^2(k+p-q-\nu,\nu,p,q)]\\
&\times[(\bar n_q-\bar n_p)\bar N_\nu\bar N_{q-\nu-p-k}-\bar n_p(1+\bar n_q)(1+\bar N_\nu+N_{q-\nu-p-k})]\\
&-[T(-k,\nu,p,q)T(-k,k+p-q-\nu,p,q)+T(k+p-q-\nu,\nu,p,q)\\
&\times T(k+p-q-\nu,k+p-q-\nu,p,q)][\bar n_q(1+\bar n_p)\bar N_{q-k+p+\nu}\\
&+\bar n_p(1+\bar n_q)(1+N_{q-\nu-p-k})]\}\delta(E_p-E_q-\bar\omega_\nu+\bar\omega_{q-\nu-p-k}+\bar\omega_k)\\
&+\frac{\pi}{N^3}\sum_{q,p,\nu}[T^2(-k,\nu,p,q)+T^2(k+p-q-\nu,\nu,p,q)]\langle S_p^zS_{-p}^z\rangle\langle S_q^zS_{-q}^z\rangle\\
&\times\{(1+\bar N_\nu+\bar N_{k+p-q-\nu})[\delta(E_p-E_q-\bar\omega_\nu-\bar\omega_{k+p-q-\nu}+\bar\omega_k)\\
&-\delta(E_p-E_q-\bar\omega_\nu+\bar\omega_{q-\nu-p-k}+\bar\omega_k)]+(\bar N_\nu-\bar N_{k+p-q-\nu})\\
&\times[\delta(E_p-E_q+\bar\omega_\nu-\bar\omega_{\nu+p+k-q}+\bar\omega_k)\\
&-\delta(E_p-E_q-\bar\omega_\nu+\bar\omega_{q+\nu-p-k}+\bar\omega_k)]\}\\
&+\frac{\pi}{N^3}\sum_{q,p,\nu}[T(-k,\nu,p,q)T(-k,k+p-q-\nu,p,q)\\
&+T(k+p-q-\nu,\nu,p,q)T(k+p-q-\nu,k+p-q-\nu,p,q)]\\
&\times\langle S_p^zS_{-p}^z\rangle\langle S_q^zS_{-q}^z\rangle(1+2\bar N_{p+k-q-\nu})[\delta(E_p-E_q-\bar\omega_\nu-\bar\omega_{k+p-q-\nu}+\bar\omega_k)\\
&-\delta(E_p-E_q-\bar\omega_\nu+\bar\omega_{q-\nu-p-k}+\bar\omega_k)].
\end{aligned}
\tag{24}
$$

$\gamma_{ph-ph}(k)$ is the phonon damping due to the phonon–phonon anharmonic interaction:

$$
\begin{aligned}
\gamma_{ph-ph}(k)=&\frac{3\pi}{N}\sum_q[B^2(q,-k,k-q)+B^2(q,k-q,-k)](\bar N_q-\bar N_{k-q})\\
&\times[\delta(\bar\omega_k-\bar\omega_q-\bar\omega_{k-q})+\delta(\bar\omega_k-\bar\omega_q+\bar\omega_{q-k})]\\
&+\frac{8\pi}{N^2}\sum_{q,p}[A^2(q,-k,p,k-q-p)+A^2(q,p,-k,k-q-p)]\\
&\times[\bar N_p(1+\bar N_q+\bar N_{p+k-q})-\bar N_q\bar N_{p+k-q}]\delta(\bar\omega_k-\bar\omega_q+\bar\omega_p-\bar\omega_{k+p-q})
\end{aligned}
\tag{25}
$$

where

$$
\langle S^z\rangle=\frac{1}{N}\sum_k\left[(S+\tfrac12)\coth\left(\frac{(S+\frac12)E_k}{k_BT}\right)-\tfrac12\coth\left(\frac{E_k}{2k_BT}\right)\right]
\tag{26}
$$

$$
\bar n_q=\langle S_q^-S_q^+\rangle/2\langle S^z\rangle\{(\varepsilon_q^{11}/E_q)\coth[E_q/2k_BT]-1\}.
\tag{27}
$$

E_q and ε_q^{11} are the spin-wave energy and one of its matrix elements, respectively, which will be calculated in the next section.

At $T = 0$, where $\gamma_{ph-ph}(k)$ vanishes, we obtain

$$\begin{aligned}\gamma^{ph}(k; T = 0) = \frac{\pi \langle S_z\rangle^4}{N^2} \sum_{q,p} &[R^2(-k, p, q) + R^2(k - q + p, p, q)] \\ &\times[\delta(E_p - E_q - \bar{\omega}_{k+p-q} + \bar{\omega}_k) - \delta(E_p - E_q + \bar{\omega}_{q-k-p} + \bar{\omega}_k)]\delta_{q0}\delta_{p0} \\ &+\frac{\pi \langle S_z\rangle^4}{N^3} \sum_{q,p,\nu} \{T(-k, \nu, p, q)[T(-k, \nu, p, q) + T(-k, k + p - q - \nu, p, q)] \\ &+T(k + p - q - \nu, \nu, p, q)[T(k + p - q - \nu, \nu, p, q) \\ &+T(k + p - q - \nu, k + p - q - \nu, p, q)]\} \\ &\times[\delta(E_p - E_q - \bar{\omega}_\nu - \bar{\omega}_{k+p-q-\nu} + \bar{\omega}_k) \\ &-\delta(E_p - E_q - \bar{\omega}_\nu + \bar{\omega}_{q-\nu-p-k} + \bar{\omega}_k)]\delta_{q0}\delta_{p0}. \end{aligned} \quad (28)$$

It is seen that at $T = 0$ the phonon modes are damped due to the spin–phonon interaction if the δ-function can be satisfied. Only the spin–phonon anharmonic terms contribute to $\gamma^{ph}(k)$ at $T = 0$.

The expression for the damping at $T \geqslant T_C$ is $\gamma^{ph}(k, T \geqslant T_{\mathrm{C}}) = \gamma_{ph-ph}(k)$, because $\gamma_{sp}(k, T \geqslant T_{\mathrm{C}}) = 0$, 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. $\langle S^z_q S^z_{-q}\rangle \to \langle S^z_0\rangle^2\delta_{q0}$. If we take into account these correlation functions we would obtain a finite contribution from the spin–phonon interaction, i.e. $\gamma_{sp}(k; T \geqslant T_{\mathrm{C}}) \neq 0$.

The phonon damping $\gamma^{ph}(k)$ was calculated numerically using the same parameters as for $\bar{\omega}_k$ for the A mode. Figure 5 shows the temperature dependence of the phonon damping. The damping is extremely small at low temperatures. Approaching T_C, $\gamma^{ph}(k)$ increases. The calculated curve is sufficiently consistent with the experimental curve for the A mode presented by Wakamura and Arai [2]. It has been shown that the spin–phonon anharmonic terms play an important role at low temperatures, whereas the anharmonic phonon–phonon interaction is important at $T \geqslant T_C$.

4. The spin-wave spectrum

4.1. The spin-wave theory

For the spin-wave energy E_k in the generalized Hartree–Fock approximation we obtain

$$E_{1/2}(k) = 0.5\{(\varepsilon^{11}_k + \varepsilon^{22}_k) \pm [(\varepsilon^{11}_k - \varepsilon^{22}_k)^2 + 4\varepsilon^{12}_k\varepsilon^{21}_k]^{1/2}\} \quad (29)$$

where

$$\begin{aligned}\varepsilon^{11}_k = g\mu_B\mathbb{H} + \frac{1}{2\langle S^z\rangle}\Bigg(&\frac{1}{N}\sum_q (J_q - J_{k-q})(2\langle S^z_q S^z_{-q}\rangle + \langle S^-_q S^+_q\rangle) \\ &+\frac{I}{N^2}\sum_{q,p}(\langle S^-_{p-q}c^+_{p+}c_{q-}\rangle + \langle S^z_{q-p}c^+_{p+}c_{q+}\rangle - \langle S^z_{q-p}c^+_{p-}c_{q-}\rangle) \\ &+\frac{1}{N}\sum_{q,p}[F_{kpq}\langle Q_{k-p-q}\rangle(2\langle S^z_q S^z_{p-k}\rangle - \langle S^-_{k-q}S^+_p\rangle) + F(p, q)\langle Q_{p-q}\rangle\langle S^z_{q-p}\rangle] \\ &+\frac{1}{2N}\sum_{q,p,\nu}[R_{\nu kpq}\langle Q_\nu Q_{k-p-q-\nu}\rangle(2\langle S^z_q S^z_{p-k}\rangle - \langle S^-_{k-q}S^+_p\rangle) \\ &+R(\nu, p, q)\langle Q_\nu Q_{p-q-\nu}\rangle\langle S^z_{q-p}\rangle] \\ &+\frac{1}{3N}\sum_{q,p,\nu,\mu}[T_{\mu\nu kpq}\langle Q_\mu Q_\nu Q_{k-p-q-\nu-\mu}\rangle(2\langle S^z_q S^z_{p-k}\rangle - \langle S^-_{k-q}S^+_p\rangle) \end{aligned}$$

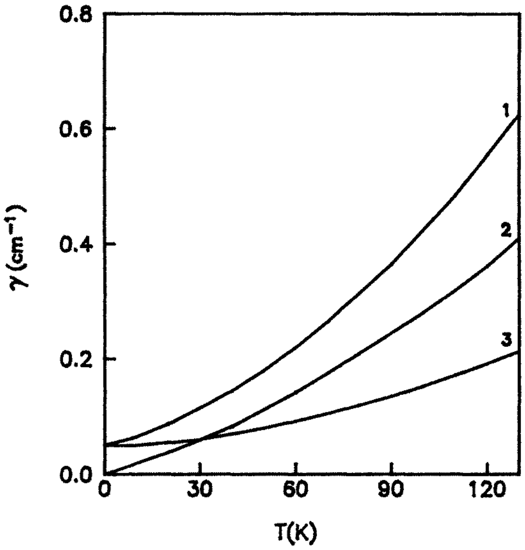

Figure 5. Temperature dependence of phonon damping with $\omega_0 = 381$ cm^{-1} and $R > 0$: curve 1, total phonon damping $\gamma^{ph}(k)$; curve 2, phonon damping $\gamma_{ph-ph}(k)$ from the anharmonic phonon–phonon interaction; curve 3, phonon damping $\gamma_{sp}(k)$ due to the s–p interaction.

$$+T(\mu, \nu, p, q)\langle Q_\mu Q_\nu Q_{p-q-\nu-\mu}\rangle\langle S^z_{q-p}\rangle]\Big) \tag{30}$$

$$\varepsilon_k^{12} = \frac{I}{2\rho N^2}\bigg(\sum_{q,p}(\langle S^z_{k-q+p}c^+_{p-k-}c_{q-}\rangle - \langle S^z_{k-q+p}c^+_{p+}c_{q+k+}\rangle)$$

$$-\tfrac{1}{2}\sum_{q,p}(\langle S^+_{k+q-p}c^+_{p-k-}c_{q+}\rangle - \langle S^+_{k+q-p}c^+_{p-}c_{q+k+}\rangle\bigg) \tag{31}$$

$$\varepsilon_k^{21} = \frac{I}{2\langle S^z\rangle N^2}\bigg(\sum_{q,p}(\langle S^z_{q-p-k}c^+_{p-}c_{q-k-}\rangle - \langle S^z_{q-p-k}c^+_{p+k+}c_{q+}\rangle)$$

$$-\tfrac{1}{2}\sum_{q,p}(\langle S^-_{k-q+p}c^+_{p+}c_{q-k-}\rangle - \langle S^+_{k-q+p}c^+_{p+k+}c_{q-}\rangle)\bigg) \tag{32}$$

$$\varepsilon_k^{22} = 2\mu_B\mathbb{H} + \frac{I}{2\rho N^2}\sum_{q,p}(\langle S^+_{p-q}c^+_{p-}c_{q+}\rangle + \langle S^z_{q-p}c^+_{p+}c_{q+}\rangle - \langle S^z_{q-p}c^+_{p-}c_{q-}\rangle) \tag{33}$$

where

$$F_{kpq} = F(k-p, q) - F(p, k-q)$$
$$R_{\nu kpq} = R(\nu, k-p, q) - R(\nu, p, k-q)$$
$$T_{\mu\nu kpq} = T(\mu, \nu, k-p, q) - T(\mu, \nu, p, k-q).$$

The matrix elements ε_k^{ij} of the spin-wave energy below T_C, when we neglect the transverse correlation function $\langle S_q^- S_q^+\rangle$ and decouple the longitudinal correlation function

$\langle S_q^z S_{-q}^z \rangle \rightarrow \langle S^z \rangle \delta_{q0}$ are (RPA)

$$\begin{aligned} \varepsilon_k^{11} &= g\mu_B \mathbb{H} + \langle S^z \rangle (J_{eff} - J_k) + I\rho \\ \varepsilon_k^{12} &= -I\langle S^z \rangle \\ \varepsilon_k^{21} &= -I\rho \\ \varepsilon_k^{22} &= 2\mu_B \mathbb{H} + I\langle S^z \rangle \end{aligned} \tag{34}$$

where

$$J_{eff} = J_0 + \tfrac{1}{2} F_k \langle Q_k \rangle + \frac{1}{2N} \sum_\nu R_{\nu k}(2\bar{N}_\nu + 1) + \frac{1}{2N} \sum_\nu T_{\nu k}(2\bar{N}_\nu + 1)\langle Q_k \rangle \tag{35}$$

with $\langle Q_k \rangle$ from equation (20).

Therefore the anharmonic spin–phonon interaction causes a renormalization of the spin–spin interaction constant $J_0 \rightarrow J_{eff}$ below T_C. Now J_{eff} is temperature dependent.

ρ is the conduction-electron magnetization and is given by

$$\rho = \frac{n_+ - n_-}{2N} = \frac{1}{2N} \sum_{q\sigma} \sigma \langle c_{q\sigma}^+ c_{q\sigma} \rangle \tag{36}$$

where n_+ and n_- are the numbers of conduction electrons in spin-up and spin-down bands, respectively. In order to calculate ρ, it is necessary to define a one-electron Green function by $G_\sigma(k) = \langle\langle c_{k\sigma}^+ c_{k\sigma} \rangle\rangle$. The electron energy is obtained as

$$\xi_\sigma(k) = \xi_k^0 - \mu - \sigma(\mu_B \mathbb{H} - 0.5I\langle S^z \rangle) \tag{37}$$

where ξ_k^0 is the conduction band energy in the paramagnetic state and μ is the chemical potential. For a simple-cubic lattice and next-neighbour interaction ξ_k^0 is given by

$$\xi_k^0 = -\frac{W}{3}[\cos(k_x a) + \cos(k_y a) + \cos(k_z a)]$$

where W is the conduction band width.

For the electron correlation function we have

$$\bar{m}_{q\sigma} = \langle c_{q\sigma}^+ c_{q\sigma} \rangle = 1/[\exp(\xi_{q\sigma}/k_B T) + 1]. \tag{38}$$

The spin-wave energy was numerically calculated taking parameters for $CdCr_2Se_4$. Figure 6 demonstrates the temperature dependence of the spin-wave energy; curve 2 shows $E_+(T)$ without the spin–phonon interaction, and curves 1 and 3 show $E_+(T)$ with the anharmonic spin–phonon interaction for $R < 0$ and $R > 0$, respectively, included. If we take into account the scattering of the spin excitation from the phonons, keeping not only the one-phonon influence, we see that the phase transition temperature T_C is changed.

The temperature dependences of J_{eff} for different R-values are plotted in figure 7. The effective spin–spin interaction displays a non-linear temperature dependence. One may expect that the consideration of the spin–phonon interaction will lead to a considerable change in T_C, but we do not observe that in figure 6. The reason is the s–d interaction. In our real system, $I \gg J_{eff}$. Theoretically the change in J_0 is an effect which is due to the higher-order perturbation in the spin-wave energy and this does not change the values of $\langle S^z \rangle$ and T_C considerably.

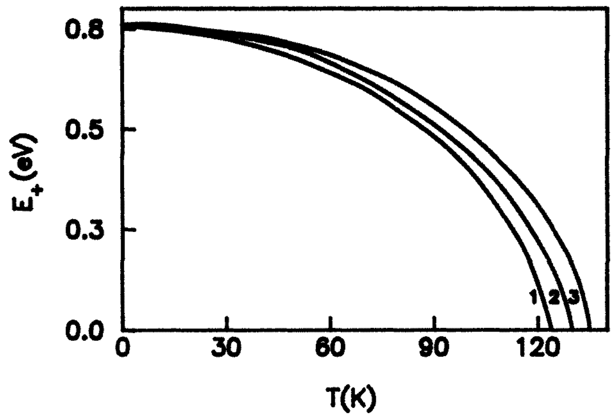

Figure 6. Temperature dependence of the spin-wave energy $E_+(k)$ for $R < 0$ (curve 1), $R = 0$ (curve 2) and $R > 0$ (curve 3).

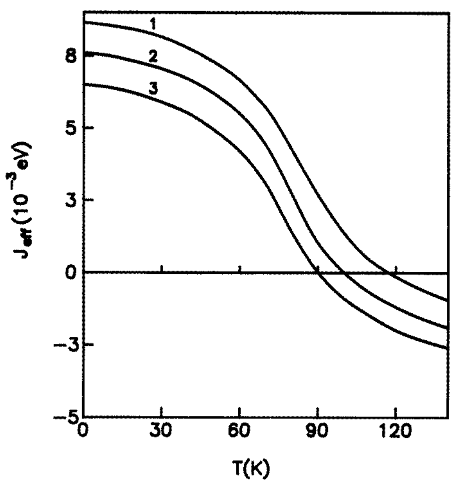

Figure 7. Temperature dependence of the effective spin–spin interaction $J_{eff}(T)$ for different values of R: curve 1, $R = 2$ cm^{-1}; curve 2, $R = -3$ cm^{-1}; curve 3, $R = -18$ cm^{-1}.

4.2. Spin-wave damping

In order to obtain spin-wave damping caused by the spin–phonon interaction, we consider the integral term (11). In our calculations we use the approximate dynamics

$$S_k(t) \sim S_k \exp(-\mathrm{i}E_k t) \qquad c_{k\sigma}(t) \sim c_{k\sigma} \exp(-\mathrm{i}\xi_{k\sigma}) \qquad a_k(t) \sim a_k \exp(-\mathrm{i}\bar{\omega}_k t)$$

where E_k, $\xi_{k\sigma}$ and $\bar{\omega}_k$ come from (29), (37) and (14), respectively. This assumption takes the generalized Hartree–Fock approximation as a starting approximation.

Calculations yield the following expression for the transverse damping $\gamma^s(k)$:

$$\gamma^s(k) = \gamma_{ss}(k) + \gamma_{sd}(k) + \gamma_{sp}(k). \tag{39}$$

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

$$\gamma_{ss}(k) = \frac{2\pi\langle S^z\rangle^2}{N^2}\sum_{q,p} V_{kqp}^2[\bar{n}_p(1+\bar{n}_{k-q}+\bar{n}_{p+q}) - \bar{n}_{k-q}\bar{n}_{p+q}]\delta(E_{p+q}+E_{k-q}-E_p-E_k) \tag{40}$$

where $V_{kqp} = (J_q + J_{k-q-p}) - (J_{k-q} + J_{p+q})$.

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

$$\begin{aligned}\gamma_{sd}(k) = {} & \frac{2\pi I^2\langle S^z\rangle}{N^3}\sum_{q,p,r}[(\bar{n}_p - \bar{n}_{p+k+q})\bar{m}_{q+r+}(1-\bar{m}_{r-}) + \bar{n}_{p+k+q}(1+\bar{n}_p)(\bar{m}_{q+r+} - \bar{m}_{r-})] \\ & \times\delta(E_{p+k+q} - E_p + \xi_{q+r+} - \xi_{r-} - E_k) \\ & + \frac{\pi I^2}{4N^2}\sum_{q,p,\sigma}[\bar{m}_{p+q\sigma}(1-\bar{m}_{p\sigma}) + \bar{n}_{k-q}(\bar{m}_{p+q\sigma} - \bar{m}_{p\sigma})] \\ & \times\delta(E_{k-q} + \xi_{p+q\sigma} - \xi_{p\sigma} - E_k) \\ & + \frac{\pi I^2\langle S^z\rangle}{2N}\sum_q(\bar{m}_{q-k+} - \bar{m}_{q-})\delta(\xi_{q-k+} - \xi_{q-} - E_k).\end{aligned} \tag{41}$$

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

$$\begin{aligned}\gamma_{sp}(k) = {} & \frac{2\pi\langle S^z\rangle^2}{N^3}\sum_{q,p,r}\{F_{kqpr}^2[\bar{n}_r(1+\bar{n}_{q+r}+\bar{n}_p) - \bar{n}_{q+r}\bar{n}_p] \\ & \times[(1+\bar{N}_{k-p-q})\delta(E_{q+r} - E_r + E_p - \bar{\omega}_{k-p-q} - E_k) \\ & + \bar{N}_{k-p-q}\delta(E_{q+r} - E_r + E_p + \bar{\omega}_{k-p-q} - E_k)] \\ & + F_{kqpr}^2(1+\bar{n}_p)(1+\bar{n}_{q+r})\bar{n}_r[\delta(E_{q+r} - E_r + E_p - \bar{\omega}_{k-p-q} - E_k) \\ & - \delta(E_{q+r} - E_r + E_p + \bar{\omega}_{k-p-q} - E_k)]\} \\ & + \frac{\pi}{4N}\sum_q F_{kq}^2[(1+\bar{N}_{q-k}+\bar{n}_q)\delta(E_q - \bar{\omega}_{q-k} - E_k) \\ & + (\bar{N}_{q-k} - \bar{n}_q)\delta(E_q + \bar{\omega}_{q-k} - E_k)] \\ & + \frac{\pi}{2N}\sum_{q,p,\nu}\{R^2(\nu,p,q)[[(1+\bar{N}_{p-q-\nu})(1+\bar{N}_\nu) + \bar{N}_\nu n_{k+q-p}] \\ & \times\delta(E_{k+q-p} + \bar{\omega}_\nu + \bar{\omega}_{p-q-\nu} + E_k) \\ & + [\bar{N}_\nu\bar{N}_{q+\nu-p} - (1+\bar{N}_{q+\nu-p} + \bar{N}_\nu)\bar{n}_{k+q-p}]\delta(E_{k+q-p} - \bar{\omega}_\nu - \bar{\omega}_{q+\nu-p} + E_k)] \\ & + R(\nu,p,q)R(p-q-\nu,p,q)[(1+\bar{N}_{p-q-\nu})(1+\bar{n}_{k+q-p}) \\ & \times\delta(E_{k+q-p} + \bar{\omega}_\nu + \bar{\omega}_{p-q-\nu} + E_k) - [\bar{N}_{q+\nu-p} + (1+\bar{N}_{q+\nu-p})\bar{n}_{k+q-p}] \\ & \times\delta(E_{k+q-p} - \bar{\omega}_\nu - \bar{\omega}_{q+\nu-p} + E_k)]\} \\ & + \frac{\pi}{2N}\sum_{q,p,\nu} R^2(\nu,p,q)[(1+\bar{N}_{q+\nu-p})\bar{N}_\nu + (\bar{N}_{q+\nu-p} - \bar{N}_\nu)\bar{n}_{k+q-p}] \\ & \times[\delta(E_{k+q-p} + \bar{\omega}_\nu - \bar{\omega}_{q+\nu-p} + E_k) + \delta(E_{k+q-p} - \bar{\omega}_\nu + \bar{\omega}_{p-q-\nu} + E_k)] \\ & + \frac{4\pi\langle S^z\rangle^2}{N^3}\sum_{q,p,\nu,r}\{R_{\nu kpqr}^2[\bar{n}_r(1+\bar{n}_{q+r}+\bar{n}_p) - \bar{n}_{q+r}\bar{n}_p][(1+\bar{N}_{k-q-p-\nu} + \bar{N}_\nu) \\ & \times\delta(E_{q+r} - E_r + E_p + \bar{\omega}_\nu + \bar{\omega}_{k-p-q-\nu} - E_k) \\ & + (\bar{N}_{q+p+\nu-k} + \bar{N}_\nu)\delta(E_{q+r} - E_r + E_p - \bar{\omega}_\nu - \bar{\omega}_{p+q+\nu-k} - E_k)] \\ & + R_{\nu kpqr}^2(1+\bar{n}_p)(1+\bar{n}_{q+r})\bar{n}_r(\bar{N}_{q+p+\nu-k} + \bar{N}_\nu)[\delta(E_{q+r} - E_r + E_p + \bar{\omega}_\nu \\ & - \bar{\omega}_{p+q+\nu-k} - E_k) - \delta(E_{q+r} - E_r + E_p - \bar{\omega}_\nu + \bar{\omega}_{k-p-q-\nu} - E_k)]\} \\ & + \frac{4\pi\langle S^z\rangle^2}{N^3}\sum_{q,p,\nu,r} R_{k-q-p-\nu\nu pqr}^2[\bar{n}_r(1+\bar{n}_{q+r}+\bar{n}_p) - \bar{n}_{q+r}\bar{n}_p](1+\bar{N}_{k-q-p-\nu}) \\ & \times[\delta(E_{q+r} - E_r + E_p + \bar{\omega}_\nu + \bar{\omega}_{k-p-q-\nu} - E_k)\end{aligned}$$

$$
\begin{aligned}
&-\delta(E_{q+r}-E_r+E_p-\bar{\omega}_\nu-\bar{\omega}_{p+q+\nu-k}-E_k)]\\
&+\frac{\pi}{2N}\sum_{q,p,\nu,\mu}\{T^2(\mu,\nu,p,q)[[(1+\bar{N}_{p-q-\mu-\nu})(1+\bar{N}_\nu)(1+\bar{N}_\mu)\\
&+\bar{N}_{p-q-\mu-\nu}(1+\bar{N}_\nu+\bar{N}_\mu)\bar{n}_{k+q-p}]\\
&\times\delta(E_{k+q-p}+\bar{\omega}_\mu+\bar{\omega}_\nu+\bar{\omega}_{p-q-\nu-\mu}-E_k)\\
&+[\bar{N}_{q+\nu+\mu-p}\bar{N}_\nu(1+\bar{N}_\mu+\bar{n}_{k+q-p})-\bar{N}_\mu(1+\bar{N}_{q+\nu+\mu-p}+\bar{N}_\nu)\\
&\times\delta(E_{k+q-p}-\bar{\omega}_\mu-\bar{\omega}_\nu-\bar{\omega}_{q+\nu+\mu-p}-E_k)]\\
&+T(\mu,\nu,p,q)T(\mu,p-q-\mu-\nu,p,q)[[(1+\bar{N}_\mu)(1+\bar{N}_{p-q-\mu-\nu})\\
&+\{(1+\bar{N}_\nu)(1+\bar{N}_{p-q-\mu-\nu})+\bar{N}_\nu\bar{N}_{p-q-\mu-\nu}\}\bar{n}_{k+q-p}]\\
&\times\delta(E_{k+q-p}+\bar{\omega}_\mu+\bar{\omega}_\nu+\bar{\omega}_{p-q-\nu-\mu}-E_k)\\
&-[\bar{N}_\mu\bar{N}_{q+\nu+\mu-p}+\bar{N}_\mu(1+\bar{N}_{q+\nu+\mu-p})\bar{n}_{k+q-p}]\\
&\times\delta(E_{k+q-p}-\bar{\omega}_\mu-\bar{\omega}_\nu-\bar{\omega}_{q+\nu+\mu-p}-E_k)]]\}\\
&+\frac{\pi}{3N}\sum_{q,p,\nu,\mu}T^2(\mu,\nu,p,q)[(1+\bar{N}_{q+\mu+\nu-p})(1+\bar{N}_\nu)\bar{N}_\mu\\
&+\{\bar{N}_{q+\mu+\nu-p}(1+\bar{N}_\nu+\bar{N}_\mu)-\bar{N}_\nu\bar{N}_\mu\}\bar{n}_{k+q-p}]\\
&\times[\delta(E_{k+q-p}+\bar{\omega}_\mu+\bar{\omega}_\nu-\bar{\omega}_{q+\nu+\mu-p}-E_k)\\
&+\delta(E_{k+q-p}-\bar{\omega}_\mu-\bar{\omega}_\nu+\bar{\omega}_{p-q-\nu-\mu}-E_k)]\\
&+\frac{\pi\langle S^z\rangle^2}{3N^3}\sum_{q,p,\nu,\mu,r}\{T^2_{\mu\nu kqpr}[\bar{n}_r(1+\bar{n}_{q+r}+\bar{n}_p)-\bar{n}_{q+r}\bar{n}_p]\\
&\times[\{\bar{N}_\nu\bar{N}_\mu+(1+\bar{N}_\nu+\bar{N}_\mu)(1+\bar{N}_{k-p-q-\nu-\mu})\}\\
&\times\delta(E_{r+q}-E_r+E_p+\bar{\omega}_\nu+\bar{\omega}_\mu+\bar{\omega}_{k-p-q-\nu-\mu}-E_k)+\bar{N}_{p+q+\nu+\mu-k}\\
&\times(1+\bar{N}_\nu+\bar{N}_\mu)\delta(E_{r+q}-E_r+E_p-\bar{\omega}_\nu-\bar{\omega}_\mu-\bar{\omega}_{p+q+\nu+\mu-k}-E_k)]\\
&+T^2_{\mu\nu kqpr}(1+\bar{n}_p)(1+\bar{n}_{q+r})\bar{n}_r[\bar{N}_{p+q+\nu+\mu-k}(1+\bar{N}_\nu+\bar{N}_\mu)+\bar{N}_\nu\bar{N}_\mu]\\
&\times[\delta(E_{r+q}-E_r+E_p+\bar{\omega}_\nu+\bar{\omega}_\mu-\bar{\omega}_{p+q+\nu+\mu-k}-E_k)\\
&-\delta(E_{r+q}-E_r+E_p-\bar{\omega}_\nu-\bar{\omega}_\mu+\bar{\omega}_{k-p-q-\nu-\mu}-E_k)]\}\\
&+\frac{\pi\langle S^z\rangle^2}{3N^3}\sum_{q,p,\nu,\mu,r}T^2_{\mu k-p-q-\mu kpqr}[\bar{n}_r(1+\bar{n}_{q+r}+\bar{n}_p)-\bar{n}_{q+r}\bar{n}_p]\\
&\times\bar{N}_\mu(1+N_{p+q+\nu+\mu-k})[\delta(E_{r+q}-E_r+E_p+\bar{\omega}_\nu+\bar{\omega}_\mu+\bar{\omega}_{k-p-q-\nu-\mu}-E_k)\\
&-\delta(E_{r+q}-E_r+E_p-\bar{\omega}_\nu-\bar{\omega}_\mu-\bar{\omega}_{p+q+\nu+\mu-k})]
\end{aligned}
\tag{42}
$$

where

$$
\begin{aligned}
F_{kqpr}&=[F(q,k-p)+F(p-r,k-q-r)]\\
&\quad-[F(k-q,p)+F(k-p+r,q+r)]\\
R_{\nu kqpr}&=[R(\nu,q,k-p)+R(\nu,p-r,k-q-r)]\\
&\quad-[R(\nu,k-q,p)+R(\nu,k-p+r,q+r)]\\
T_{\mu\nu kqpr}&=[T(\mu,\nu,q,k-p)+T(\mu,\nu,p-r,k-q-r)]\\
&\quad-[T(\mu,\nu,k-q,p)+T(\mu,\nu,k-p+r,q+r)].
\end{aligned}
$$

At $T=0$ we obtain for $\gamma^s(k,T=0)$

$$
\begin{aligned}
\gamma^s(k)=\frac{\pi}{4N}&\sum_q F^2_{kq}\delta(E_q-\bar{\omega}_{q-k}-E_k)\\
&+\frac{\pi}{2N}\sum_{q,p,\nu}[R^2(\nu,p,q)+R(\nu,p,q)R(p-q-\nu,p,q)]\\
&\times\delta(E_{q+r}-E_r+E_p+\bar{\omega}_\nu+\bar{\omega}_{k-p-q-\nu}-E_k)\\
&+\frac{\pi}{3N}\sum_{q,p,\nu,\mu}[T^2(\mu,\nu,p,q)+T(\mu,\nu,p,q)T(\mu,p-q-\mu-\nu,p,q)]\\
&\times\delta(E_{r+q}-E_r+E_p+\bar{\omega}_\nu+\bar{\omega}_\mu+\bar{\omega}_{k-p-q-\nu-\mu}-E_k).
\end{aligned}
\tag{43}
$$

We can see that at $T = 0$ the spin waves are damped due to the spin–phonon interaction if the δ-function can be satisfied.

At $T \geqslant T_C$, $\gamma_{ss}(k) = 0$, i.e. the spin-wave damping in the high-temperature region is due to the s–d and s–p interaction.

The temperature dependence of the spin-wave damping is presented in figure 8. At low temperatures the damping is very small, the spin-wave energy E_k is greater than the damping term. Approaching T_C, $\gamma^s(k)$ 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

$$\gamma_{ss}(k) \ll \gamma_{sp}(k) \leqslant \gamma_{sf}(k). \tag{44}$$

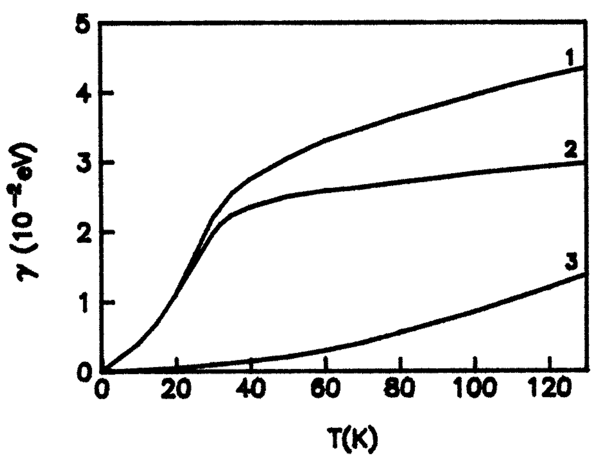

Figure 8. Temperature dependence of spin-wave damping with $F = 23$ cm^{-1}, $R = -18$ cm^{-1}, $T = 1.4$ cm^{-1} and $\omega_0 = 100$ cm^{-1}: curve 1, total spin-wave damping $\gamma^s(k)$; curve 2, spin-wave damping $\gamma_{sd}(k)$ from the s–d interaction; curve 3, the spin-wave damping $\gamma_{sp}(k)$ due to the s–p interaction.

5. Conclusions

The present paper presents a study of the anharmonic effects in ferromagnetic semiconductors. It has been found that the anharmonic spin–phonon and phonon–phonon interactions modify the initial phonon frequency. The temperature dependences of two modes with $\omega_0 = 100$ cm^{-1} and $\omega_0 = 381$ cm^{-1} have been calculated numerically. These modes display a non-linear temperature dependence. At $T \leqslant T_C$ this behaviour can be explained as 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. If we take into account only the third-order phonon–phonon interaction, we obtain a linear temperature dependence. The theoretical results are in very good agreement with the experimental data which were published by Wakamura and Arai [2]. The renormalized spin-wave energy has been obtained too. The spin–phonon interaction causes a renormalization of the spin–spin interaction constant $J_0 \to J_{eff}$, which is now temperature dependent. The spin–phonon interaction has renormalized the phase transition temperature T_C. Phonon damping and spin–wave damping have been evaluated, taking into account higher-order anharmonic terms, and have been numerically calculated using model parameters for $CdCr_2Se_4$. The influences of the s–s, s–d and s–p interactions on damping have been discussed. 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 $T \geqslant T_C$. The results for damping are in agreement with the experimental data [2, 11].

In conclusion it has to be noted that correct results for the spin-wave energy and phonon energy and also for spin-wave damping and phonon damping in ferromagnetic semiconductors require the consideration of the third- and fourth-order anharmonic terms in the spin–phonon and phonon–phonon interactions.

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References

[1] Guntherodt G and Zeyher R 1984 *Light Scattering in Solids* vol 4, ed M Cardona and G Guntherodt (Berlin: Springer) p 203
[2] Wakamura K and Arai T 1990 *Phase Trans.* **27** 129
[3] Sintani K, Tsuchida A and Siratori K 1968 *J. Phys. Soc. Japan* **25** 99
[4] Wakamura K and Arai T 1988 *J. Appl. Phys.* **63** 5824
[5] Kashchev V and Krivoglaz M 1961 *Fiz. Tverd. Tela* **3** 1117
[6] Yakovlev E 1962 *Fiz. Tverd. Tela* **4** 433
[7] Pytte E 1965 *Ann. Phys., NY* **32** 377
[8] Konwent H 1968 *Phys. Lett.* **28A** 237
[9] Gluck P and Entin-Wohlman O 1972 *Phys. Status Solidi* b **52** 323
[10] Wesselinowa J and Apostolov A 1993 *J. Phys.: Condens. Matter* **5** 3555
[11] Anisimov A and Green J 1983 *Zh. Eksp. Teor. Fiz.* **84** 1407
[12] Gurevich A and Anisimov A 1985 *Acta Phys. Pol.* A **68** 467
[13] Tserkovnikov Yu 1971 *Theor. Math. Phys.* **7** 250
[14] Kanamata T and Shivakama K 1985 *J. Phys. Soc. Japan* **54** 334
[15] Sakai N and Pifer J 1986 *Phys. Rev.* B **33** 1875