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LETTER TO THE EDITOR

Effects of magnetic ordering on the phonon damping in ferromagnetic semiconductors

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Abstract. A Green function technique is developed to study the spin–phonon interaction's influence on the phonon spectrum and phonon damping in ferromagnetic semiconductors. The temperature dependence of these quantities is discussed, and is found to be in agreement with the experimental data.

The effects of magnetic ordering on phonon modes have been mainly investigated from Raman spectra in Eu chalcogenides [1]. A spin-dependent Raman band was observed below the Curie temperature T_C . It was assigned to be due to the phonons at the boundary of the Brillouin zone, which are forbidden above T_C but allowed below T_C by the spin disordered arrangements.

On the other hand the infrared spectra have been measured for only a few magnetic crystals. From the reststrahlen bands, the phonon parameters can be determined without considering the intermediate state that appears in Raman spectra. Therefore, the infrared measurement is appropriate for observing directly the pure effect of spin ordering on phonon modes. The rapid variations for the frequency and damping constant have been observed near T_C on the zone centre phonon of spinels (CdCr_2S_4) [2]. Recently Wakamura [3] has observed the temperature dependence of phonon damping through T_C for a ferrimagnetic semiconductor FeCr_2S_4 by measuring the infrared reflectivity spectra.

In our previous paper [4], hereafter referred to as I, we used a Green function technique to study the spin-dependent phonon Raman scattering in ferromagnetic semiconductors (FMS). A theoretical calculation of the spin–phonon interactions influence on the spin polarizability was undertaken in I. The aim of the present letter is to study theoretically, for the first time, the influence of the spin–phonon interactions on the phonon spectrum and on the phonon damping beyond the RPA.

The total Hamiltonian of the s–d model which is proposed to describe the properties of FMS including the spin–phonon interaction may be written as

$$H = H_{s-d} + H_P + H_{SP}. \quad (1)$$

Here H_{s-d} is the Hamiltonian of the s–d model [4], H_P is the usual Hamiltonian of the lattice vibrations and H_{SP} describes the interaction of the spins with the phonons,

$$H_{SP} = H_{SP}^{(1)} + H_{SP}^{(2)} = \frac{1}{2} \sum_{q,p} \bar{F}(p,q) Q_{p-q} (S_q^z S_{-p}^z + S_q^- S_p^+) + \frac{1}{4} \sum_{k,q,p} \bar{R}(k,p,q) Q_k Q_{-k+p-q} (S_q^z S_{-p}^z + S_q^- S_p^+). \quad (2)$$

Here $H_{\text{SP}}^{(1)}$ and $H_{\text{SP}}^{(2)}$ denote the spin-phonon effects arising from first and second powers in the relative displacement of lattice sites away from equilibrium. \bar{F} and \bar{R} designate the amplitudes for coupling phonons to the spin wave excitations in first and second order, respectively [4].

In order to study the influence of the spin-phonon interactions on the phonon spectrum we evaluate the Green function $G(\mathbf{k}, \omega) = \langle\langle Q_{\mathbf{k}}; Q_{-\mathbf{k}}^{\dagger} \rangle\rangle$. The vibrational normal coordinate $Q_{\mathbf{k}}$ can be expressed in terms of phonon operators, $Q_{\mathbf{k}} = (2\omega_{\mathbf{k}})^{-1/2} (a_{\mathbf{k}} + a_{-\mathbf{k}}^{\dagger})$. Using the same method as in I we obtain the renormalized phonon frequency in the generalized Hartree-Fock approximation

$$\varepsilon_{\mathbf{k}}^2 = \omega_{\mathbf{k}}^2 + \frac{2}{N} \sum_{\mathbf{q}} R(\mathbf{k}, \mathbf{q}, \mathbf{q}) (\langle S_{\mathbf{q}}^{-} S_{\mathbf{q}}^{+} \rangle + \langle S_{\mathbf{q}}^z S_{-\mathbf{q}}^z \rangle) \quad (3)$$

$$\omega_{\mathbf{k}} = v k \quad R(\mathbf{k}, \mathbf{p}, \mathbf{q}) = \bar{R}(\mathbf{k}, \mathbf{p}, \mathbf{q}) / (4\omega_{\mathbf{k}} \omega_{-\mathbf{k}+\mathbf{p}-\mathbf{q}})^{1/2}.$$

As can be seen from (3) in this approximation the second term $H_{\text{SP}}^{(2)}$ renormalizes the phonon energy. Equation (3) incorporates the spin-phonon interactions in the sense of a mean field effect. The transverse and longitudinal spin correlation functions $\langle S_{\mathbf{q}}^{-} S_{\mathbf{q}}^{+} \rangle$ and $\langle S_{\mathbf{q}}^z S_{-\mathbf{q}}^z \rangle$ were calculated in I. Above T_C the spin magnetization $\langle S^z \rangle$ vanishes. As a consequence the expression for the phonon energy is simpler than below T_C :

$$\varepsilon_{\mathbf{k}}^2 = \omega_{\mathbf{k}}^2 + \frac{2}{N} \sum_{\mathbf{q}} R(\mathbf{k}, \mathbf{q}, \mathbf{q}) \langle S_{\mathbf{q}}^z S_{-\mathbf{q}}^z \rangle \quad \text{for } T \geq T_C. \quad (4)$$

Extending the theory to higher order we obtain the expression for the phonon damping:

$$\gamma_{\text{ph}}(\mathbf{k}) = \gamma_{\text{ph}}^{(1)} + \gamma_{\text{ph}}^{(2)} \quad (5)$$

where

$$\gamma_{\text{ph}}^{(1)}(\mathbf{k}) = \frac{4\pi \langle S^z \rangle^2}{N} \sum_{\mathbf{q}} F^2(\mathbf{q}, \mathbf{q}-\mathbf{k}) (\bar{n}_{\mathbf{q}} - \bar{n}_{\mathbf{q}-\mathbf{k}}) \delta(E_{\mathbf{q}-\mathbf{k}} - E_{\mathbf{q}} + \omega_{\mathbf{k}}) \quad (6)$$

$$F(\mathbf{p}, \mathbf{q}) = \bar{F}(\mathbf{p}, \mathbf{q}) / (2\omega_{\mathbf{p}-\mathbf{q}})^{1/2}$$

and

$$\begin{aligned} \gamma_{\text{ph}}^{(2)}(\mathbf{k}) = & \frac{4\pi \langle S^z \rangle^2}{N^2} \sum_{\mathbf{q}, \mathbf{p}} \{ R^2(-\mathbf{k}, \mathbf{p}, \mathbf{q}) (\bar{n}_{\mathbf{q}} - \bar{n}_{\mathbf{p}}) [(1 + \bar{m}_{\mathbf{k}+\mathbf{p}-\mathbf{q}}) \delta(E_{\mathbf{p}} - E_{\mathbf{q}} - \omega_{\mathbf{k}+\mathbf{p}-\mathbf{q}} + \omega_{\mathbf{k}}) \\ & + \bar{m}_{\mathbf{q}-\mathbf{k}-\mathbf{p}} \delta(E_{\mathbf{p}} - E_{\mathbf{q}} + \omega_{\mathbf{q}-\mathbf{k}-\mathbf{p}} + \omega_{\mathbf{k}})] \\ & + (R^2(-\mathbf{k}, \mathbf{p}, \mathbf{q}) + R^2(\mathbf{k}-\mathbf{q}+\mathbf{p}, \mathbf{p}, \mathbf{q})) \bar{n}_{\mathbf{q}} (1 + \bar{n}_{\mathbf{p}}) \\ & \times [\delta(E_{\mathbf{q}} - E_{\mathbf{q}} - \omega_{\mathbf{k}+\mathbf{p}-\mathbf{q}} + \omega_{\mathbf{k}}) - \delta(E_{\mathbf{p}} - E_{\mathbf{q}} + \omega_{\mathbf{q}-\mathbf{k}-\mathbf{p}} + \omega_{\mathbf{k}})] \} \\ & + \frac{\pi}{N^2} \sum_{\mathbf{q}, \mathbf{p}} (R^2(-\mathbf{k}, \mathbf{p}, \mathbf{q}) + R^2(\mathbf{k}-\mathbf{q}+\mathbf{p}, \mathbf{p}, \mathbf{q})) \langle S_{\mathbf{p}}^z S_{-\mathbf{p}}^z \rangle \langle S_{\mathbf{q}}^z S_{-\mathbf{q}}^z \rangle \\ & \times [\delta(E_{\mathbf{p}} - E_{\mathbf{q}} - \omega_{\mathbf{k}+\mathbf{p}-\mathbf{q}} + \omega_{\mathbf{k}}) - \delta(E_{\mathbf{p}} - E_{\mathbf{q}} + \omega_{\mathbf{q}-\mathbf{k}-\mathbf{p}} + \omega_{\mathbf{k}})] \end{aligned} \quad (7)$$

with

$$\bar{n}_{\mathbf{q}} \equiv \langle S_{\mathbf{q}}^{-} S_{\mathbf{q}}^{+} \rangle = 1 / [\exp(E_{\mathbf{q}} / k_B T) - 1] \quad E_{\mathbf{q}} = g\mu_B H + \langle S^z \rangle (J_0 - J_{\mathbf{q}})$$

$$\bar{m}_{\mathbf{q}} \equiv \langle a_{\mathbf{q}}^{+} a_{\mathbf{q}} \rangle = 1 / [\exp(\omega_{\mathbf{q}} / k_B T) - 1] \quad \omega_{\mathbf{q}} = v q.$$

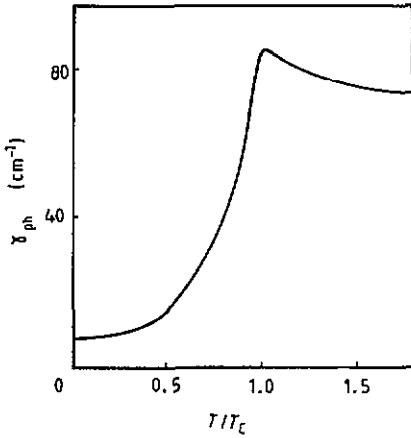


Figure 1. Temperature dependence of the phonon damping $\gamma_{\text{ph}}(T)$ for CdCr_2Se_4 .

Here $\langle S^z \rangle$ and ρ are the localized-spin magnetization and the conduction electron magnetization, respectively, which were calculated in I.

Above T_C only the last term in $\gamma_{\text{ph}}^{(2)}$ (7) remains finite due to second order spin-phonon interactions $H_{\text{SP}}^{(2)}$.

The phonon damping $\gamma_{\text{ph}}(T)$ was calculated numerically with parameters for CdCr_2Se_4 [5] ($J_0 = 0.0001$ eV, $I = 0.5$ eV, $W = 0.1$ eV, $T_C = 130$ K, $S = 7/2$), for $k = 0.2\pi, 0.2\pi, 0.2\pi$, $h \equiv g\mu_B H = 0.0001$ eV, and for different values of temperature T . The damping $\gamma_{\text{ph}}(k)$ is small at low temperatures, then increases rapidly with $T \rightarrow T_C$. Above T_C it decreases, but very slowly (figure 1). At low temperatures the two-phonon scattering processes $\gamma_{\text{ph}}^{(2)}$ give negligible contribution to the damping, whereas at higher temperatures, $T \approx T_C$ and above T_C , they give the more important contribution. It may be noted that here we have not made a $1/Z$ expansion and so we have taken into account all summation terms. The obtained behaviour of $\gamma_{\text{ph}}(T)$ is in very good agreement with the experimental results of Wakamura and Arai [2] for CdCr_2S_4 and of Wakamura [3] for FeCr_2S_4 .

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