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Effect of Magnetic Field on Sound Propagation near Magnetic Phase Transition Temperatures

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The anomalous ultrasonic attenuation and velocity variation caused by the critical fluctuation of spins near the Curie and Néel temperatures are theoretically investigated and found to be strongly affected by an application of magnetic field. In the random phase approximation, the attenuation coefficient is expressed in terms of a sum of the two terms; a cross term of the static spin polarization and the two-spin correlation function, and a product of the two-spin correlation functions. In the magnetic field, the former term has a positive contribution to the attenuation, since this term has a finite value only when the static spin polarization exists. The latter term decreases in the field, owing to the suppression of spin thermal fluctuations due to the magnetic field. The magnitude of the contributions from these terms depends upon temperature, the strength of magnetic field and the nature of the exchange interaction in magnetic materials. The theory explains various types of the field dependence of the attenuation observed in magnetic materials including MnP, Dy and MnF_2. A new attenuation peak found recently by Hirahara et al. in the paramagnetic phase of MnP under a magnetic field is explained on the basis of the present theory.

§ 1. Introduction

When the ultrasonic attenuation in magnetic materials is measured as a function of temperature, a sharp peak is usually observed at the Curie and Néel temperatures.[1] If a magnetic field is applied, the critical attenuation increases in some magnetic materials and decreases in some other magnetic materials. For example, Neighbours et al.[2] have observed a decrease of the attenuation in MnF_2 in magnetic fields, whereas Lee and Levy have found a large increase of the attenuation in Dy[3] and Ho[4] in magnetic fields. Whether the attenuation increases or decreases is also dependent on temperature. Recently, Hirahara et al.[5] have observed that the attenuation in MnP decreases in magnetic fields when temperature is extremely near the Curie point. However, the attenuation increases above a certain temperature. When temperature is fixed at some temperature above that temperature and magnetic field is applied, the attenuation starts to increase proportionally to the square of the magnetic field, attains a maximum and decreases with further increasing magnetic field. The following phenomenon which they have observed in MnP seems to be more striking. When a magnetic field is applied along the hard axis of magnetization, the Curie temperature shifts toward the lower temperature. In this case, in addition to the attenuation peak

at this shifted Curie temperature, they found a new broad peak in the temperature region where it has been considered to be a uniformly paramagnetic region.

It has been observed in magnetic materials such as Gd,[1] Dy,[6] Ho[6] and MnP[5] that the longitudinal sound velocity has a sharp dip at the critical temperature. This anomalous variation of the velocity is also strongly affected by an application of magnetic field. The aim of the present investigation is to construct a theory for the ultrasonic attenuation and the velocity change due to the spin-phonon interaction in the presence of magnetic field, and to explain systematically various types of the field effect mentioned above.

The spin-phonon interaction responsible for the critical attenuation and the change in the velocity of longitudinal sound arises in most cases from the strain modulation of the exchange interaction.[7)~11)] Via this interaction, the spin thermal fluctuation causes random forces to act on the acoustic normal modes. According to Mori's theory[12] for irreversible processes, the sound attenuation coefficient is expressed as the time-correlation function of these random forces and is proportional to the spin correlation function. In a magnetic field, the spin correlation function is divided into two parts: One is the cross product of the static spin-polarization induced by the magnetic field and the two-spin correlation function, and the other is the correlation function of four spins. The latter part corresponds to the usual mechanism[7)~11)] which has been considered for the critical attenuation in the absence of magnetic field. If a magnetic field is applied, the spin fluctuation is suppressed and the magnitude of the spin correlation function is decreased. Consequently, this part contributes to a decrease of the attenuation in the magnetic field. The first part appears only when the static spin polarization exists. Therefore, this term has a positive contribution in the presence of magnetic field in the paramagnetic phase and below the Curie temperature. This mechanism has first been introduced by Lee, Levy and one of the authors (M.T.)[4] to explain the increase of the critical attenuation in Ho in magnetic fields. The combination of the two effects mentioned above can explain various types of the field effect on the ultrasonic attenuation observed in many magnetic materials. The sound velocity as a function of temperature has a sharp dip at the Curie and Néel temperatures.[1] This anomalous variation of the velocity is also strongly affected by an application of magnetic field.[1),5),6)] This field effect is explained from a mechanism similar to that of the attenuation.

In §§ 2 and 3, expressions for the ultrasonic attenuation and the change in the sound velocity due to the spin-phonon interaction in the presence of magnetic field are obtained, using the random phase approximation. These expressions include the staggered susceptibilities. The susceptibilities in magnetic fields are calculated and inserted into these expressions, and explicit formulae for the attenuation and the velocity are obtained in § 4. The increase of the attenuation from the first mechanism mentioned above compensates the decrease from the second mechanism at a certain temperature. In § 5, it is suggested how the

compensation temperature is related to the nature of the exchange interaction. In § 6, the theory is applied to MnP, and the attenuation coefficient and the velocity change are calculated. The calculated results explain the experimental results obtained by Hirahara et al.[5] In § 7, the theory is also applied to Dy and the calculated attenuation coefficient is compared with the experimental one obtained by Lee and Levy.[3]

§ 2. Ultrasonic attenuation coefficient

For the spin-phonon interaction responsible for the critical attenuation of longitudinal sound, the strain modulations of the exchange interaction and the magnetostrictive coupling of one ion type are considered. However, in most of the magnetic materials in which the exchange energy is larger than the anisotropy energy, the contribution to the spin-phonon interaction from the exchange interaction dominates. Thus, we include only the exchange contribution in our Hamiltonian. The total Hamiltonian for a spin and acoustic phonon system is written as[8]

$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}_1+\mathcal{H}_2\,, \tag{1}$$

$$\mathcal{H}_0=\sum_{\boldsymbol{k}}\hbar\omega_{\boldsymbol{k}}{}^0 b_{\boldsymbol{k}}{}^* b_{\boldsymbol{k}}-\sum_{\alpha}\sum_{i,j}J_{ij}^{\alpha}S_i{}^{\alpha}S_j{}^{\alpha}+g\mu_{\mathrm{B}}H\sum_i S_i{}^z, \tag{2}$$

$$\mathcal{H}_1=\sum_{\boldsymbol{k}}(\hbar/2\rho V\omega_{\boldsymbol{k}}{}^0)^{1/2}(b_{\boldsymbol{k}}+b_{-\boldsymbol{k}}^*)\,U_{\boldsymbol{k}}{}^{(1)}, \tag{3}$$

$$\mathcal{H}_2=\sum_{\boldsymbol{k}}\sum_{\boldsymbol{k}'}(\hbar/2\rho V)\,(\omega_{\boldsymbol{k}}{}^0\omega_{\boldsymbol{k}'}^0)^{-1/2}(b_{\boldsymbol{k}}b_{-\boldsymbol{k}'}+b_{-\boldsymbol{k}}^*b_{\boldsymbol{k}'}^* +b_{\boldsymbol{k}}b_{\boldsymbol{k}'}^*+b_{-\boldsymbol{k}}^*b_{-\boldsymbol{k}'})\,U_{\boldsymbol{k}\boldsymbol{k}'}^{(2)}\,, \tag{4}$$

$$U_{\boldsymbol{k}}{}^{(1)}=-\sum_{i,j}(e^{i\boldsymbol{k}\cdot\boldsymbol{R}_i}-e^{i\boldsymbol{k}\cdot\boldsymbol{R}_j})\,\boldsymbol{e}_{\boldsymbol{k}}\cdot\sum_{\alpha}\frac{\partial J_{ij}^{\alpha}}{\partial\boldsymbol{R}_i}S_i{}^{\alpha}S_j{}^{\alpha}, \tag{5}$$

$$U_{\boldsymbol{k}\boldsymbol{k}'}^{(2)}=-\frac{1}{2}\sum_{i,j}(e^{i\boldsymbol{k}\cdot\boldsymbol{R}_i}-e^{i\boldsymbol{k}\cdot\boldsymbol{R}_j})\,(e^{-i\boldsymbol{k}'\cdot\boldsymbol{R}_i}-e^{-i\boldsymbol{k}'\cdot\boldsymbol{R}_j}) \times\boldsymbol{e}_{\boldsymbol{k}}\boldsymbol{e}_{-\boldsymbol{k}'}:\sum_{\alpha}\frac{\partial^2 J_{ij}^{\alpha}}{\partial\boldsymbol{R}_i\partial\boldsymbol{R}_j}S_i{}^{\alpha}S_j{}^{\alpha}, \tag{6}$$

$$\alpha=x,y,z\,,$$

where ρ is the density of crystal, V the volume of crystal, $\boldsymbol{e}_{\boldsymbol{k}}$ the polarization vector of the phonon $\boldsymbol{k}$, $\boldsymbol{R}_i$ the position vector of the i-th site, and $S_i{}^{\alpha}$ the α-component of spin at the i-th site. The first term of $\mathcal{H}_0$ represents the Hamiltonian for the non-interacting phonons whose frequencies are indicated by $\omega_{\boldsymbol{k}}{}^0$. The operators $b_{\boldsymbol{k}}$ and $b_{\boldsymbol{k}}{}^*$ are annihilation and creation boson operators, respectively. The second term represents the anisotropic exchange interaction with exchange integral J_{ij}^{α}. The third term represents the Zeeman energy in a magnetic field applied along the z axis. We assume an isotropic g-factor. The Hamiltonians $\mathcal{H}_1$ and $\mathcal{H}_2$ represent, respectively, first- and second-order spin-phonon interactions

with respect to the phonon amplitude. According to Mori,[12] the ultrasonic attenuation coefficient is expressed as a function of the time-correlation of the random force acting on the phonon b_k as[8]

$$\alpha_k = \mathrm{Re} \int_0^\infty dt\,(f_k(t), f_k^*(0)) \exp(-i\omega_k t)/v_l\,(b_k, b_k^*), \tag{7}$$

with the relaxation function defined by $(A, B) = \int_0^\beta d\lambda \langle e^{\lambda \mathcal{H}} A e^{-\lambda \mathcal{H}} B\rangle - \beta\langle A\rangle\langle B\rangle$ where $\langle C\rangle = \mathrm{Tr}\, e^{-\beta\mathcal{H}} C / \mathrm{Tr}\, e^{-\beta\mathcal{H}}$. In Eq. (7), v_l is the longitudinal sound speed and the random force is defined by

$$f_k = \dot{b}_k + i\omega_k b_k\,. \tag{8}$$

If the relation

$$\begin{aligned}\omega_k &= i(\dot{b}_k, b_k^*)/(b_k, b_k^*) = \langle [b_k, b_k^*]\rangle/\hbar(b_k, b_k^*)\\ &= 1/\hbar(b_k, b_k^*)\end{aligned} \tag{9}$$

is used, (b_k, b_k^*) is related to the phonon frequency ω_k. As will be discussed in § 3, the frequency is changed by the spin-phonon interaction. However, since this change is very small, ω_k and v_l appearing in Eq. (7) are replaced by those of non-interacting phonons. The random force f_k is expressed, from Eq. (8), as

$$f_k = -i(2\rho V\hbar\omega_k^0)^{-1/2} U_{-k}^{(1)}, \tag{10}$$

where the contribution from $\mathcal{H}_2$ was neglected, because of higher order with respect to phonon amplitude. Using Eqs. (7), (9) and (10), and introducing the Fourier amplitude of spin $\boldsymbol{S}_q = (1/N)^{1/2} \sum_i \boldsymbol{S}_i e^{i\boldsymbol{q}\cdot\boldsymbol{R}_i}$, we obtain α_k as

$$\begin{aligned}\alpha_k = (2\rho V v_l)^{-1}\,\mathrm{Re} \sum_q \sum_{q'} \sum_\alpha \sum_{\alpha'} g_q^{\alpha *}(\boldsymbol{k})\, g_{q'}^{\alpha'}(\boldsymbol{k})\\ \times \int_0^\infty (S_q^\alpha(t) S_{-q-k}^\alpha(t), S_{-q'}^{\alpha'}(0) S_{q'+k}^{\alpha'}(0)) \exp(-i\omega_k t)\, dt\,,\end{aligned} \tag{11}$$

$$g_q^\alpha(\boldsymbol{k}) = \sum_j e^{i\boldsymbol{q}\cdot\boldsymbol{R}_{ji}} (e^{i\boldsymbol{k}\cdot\boldsymbol{R}_{ji}} - 1)\, \boldsymbol{e}_k \cdot \frac{\partial J_{ij}^\alpha}{\partial \boldsymbol{R}_i}\,. \tag{12}$$

Now we introduce an external magnetic field which is applied along one of the principal axes of the exchange interaction. We take the z axis in the direction of the magnetic field. The magnetic field induces a uniform spin polarization in the z direction, and spins fluctuate around it. We divide the component S_0^z into the static spin polarization $\langle S_0^z\rangle$ and the deviation $S_0^{z\prime}$ from it

$$S_0^z = \langle S_0^z\rangle + S_0^{z\prime}. \tag{13}$$

If this expression is inserted into Eq. (11), α_k is rewritten as

$$\begin{aligned}\alpha_k = (2\rho V v_l)^{-1}\,\mathrm{Re}\Big[4 g_0^{z *}(\boldsymbol{k})\, g_0^z(\boldsymbol{k}) \langle S_0^z\rangle^2\\ \times \int_0^\infty (S_{-k}^z(t), S_k^z(0)) \exp(-i\omega_k t)\, dt + \sum_q \sum_{q'} \sum_\alpha \sum_{\alpha'} g_q^{\alpha *}(\boldsymbol{k})\, g_{q'}^{\alpha'}(\boldsymbol{k})\end{aligned}$$

$$\times \int_0^\infty (S_q^{\alpha}(t) S_{-q-k}^{\alpha}(t), S_{-q'}^{\alpha'}(0) S_{q'+k}^{\alpha'}(0)) \exp(-i\omega_k t) dt \Big], \qquad (14)$$

where S_k^{α} with $k=0$ and $\alpha=z$ in the last term reads $S_0^{z'}$. The external magnetic field induces the spin polarization and suppresses the spin thermal fluctuations. The first term in the bracket of Eq. (14) does appear only when the spin polarization exists and, thus, this term contributes to an increase of the attenuation in the presence of magnetic field. As discussed in §§ 5~7, this term plays an important role in the field dependence of the attenuation. Therefore, to elucidate the physical origin of this term, we give another derivation in the Appendix. The magnitude of the second term which is a function of four-spin correlations decreases owing to the suppression of spin fluctuations by an application of magnetic field and, thus, this term causes a decrease of the attenuation in the presence of magnetic field. For the calculation of the second term in the bracket of Eq. (14), we approximate the four-spin relaxation function by a product of the two-spin relaxation functions with the use of the decoupling approximation $(ab, cd) \sim \beta^{-1}[(a, b)(c, d) + (a, c)(b, d) + (a, d)(b, c)] - \langle ab \rangle \langle cd \rangle$. In §§ 6 and 7, we will treat spin systems in metals such as MnP and Dy. In these systems, the exchange interaction is of the long range and, therefore, the above decoupling approximation may work well except very near the transition temperature, where the correlation length of the spin pair correlation function becomes much longer than the force range of the exchange interaction. If this approximation is used, Eq. (14) is reduced to

$$\begin{aligned} \alpha_k = (2\rho V v_l)^{-1} \mathrm{Re} \Big[& 4 g_0^{z*}(k) g_0^{z}(k) \langle S_0^z \rangle^2 \int_0^\infty (S_{-k}^z(t), S_k^z(0)) \exp(-i\omega_k t) dt \\ & + \beta^{-1} \sum_q \sum_{\alpha} \sum_{\alpha'} (g_q^{\alpha *}(k) g_q^{\alpha'}(k) + g_q^{\alpha *}(k) g_{-q-k}^{\alpha'}(k)) \\ & \times \int_0^\infty (S_q^{\alpha}(t), S_{-q}^{\alpha'}(0)) (S_{-q-k}^{\alpha}(t), S_{q+k}^{\alpha'}(0)) \exp(-i\omega_k t) dt \Big]. \end{aligned} \qquad (15)$$

The next step to be done is to calculate the two-spin relaxation function in a magnetic field. For this purpose we set up the following equations of motion for S_q with damping terms:

$$\left.\begin{aligned} \dot{S}_q^x &= -\Lambda_q^y S_q^y - \Gamma_q^x S_q^x, \\ \dot{S}_q^y &= \Lambda_q^x S_q^x - \Gamma_q^y S_q^y, \\ \dot{S}_q^z &= -\Gamma_q^z S_q^z, \end{aligned}\right\} \qquad (16)$$

where

$$\left.\begin{aligned} \Lambda_q^y &= g\mu_B H + 2 \frac{\langle S_0^z \rangle}{\sqrt{N}} (J_0^z - J_q^y), \\ \Lambda_q^x &= g\mu_B H + 2 \frac{\langle S_0^z \rangle}{\sqrt{N}} (J_0^z - J_q^x), \end{aligned}\right\} \qquad (17)$$

$$J_q^{\alpha} = \sum_{(i-j)} J_{ij}^{\alpha} e^{iq\cdot(R_i - R_j)}. \tag{18}$$

In Eq. (16), Γ_q^{α} is the damping constant for the spin component S_q^{α} and is given by Mori[12] as

$$\Gamma_q^{\alpha}(z) = \mathrm{Re} \int_0^{\infty} (K_q^{\alpha}(t), K_{-q}^{\alpha}(0)) e^{-zt} dt / (S_q^{\alpha}, S_{-q}^{\alpha}), \tag{19}$$

where K_q^{α} is the random torque acting on the spin component S_q^{α}. As mentioned above, we are considering spin systems in metals such as MnP and Dy. For these spin systems, we take a model that localized spins are immersed in conduction electron sea and assume that the energy of these spin systems is dissipated by conduction electrons through the so-called *s-d* or *s-f* interaction. As shown by Kawasaki,[13] $(K_q^{\alpha}(t), K_{-q}^{\alpha}(0))$ in this case does not have anomaly at a magnetic phase transition temperature and this correlation function is expected to have a weak temperature dependence even in the vicinity of the transition temperature. Furthermore, the time-correlation of the random torque may have the short time nature and thus Γ_q^{α} becomes independent of the parameter z in Eq. (19). Therefore, in these spin systems, the conventional theory for spin damping works well and Γ_q^{α} is written as

$$\Gamma_q^{\alpha} = \beta (g\mu_B)^2 B / \chi_q^{\alpha}, \tag{20}$$

where B is a constant dependent on magnetic materials but independent of temperature, and χ_q^{α} is the staggered susceptibility per ion. In deriving Eq. (20) from Eq. (19), a relation[14]

$$(S_q^{\alpha}, S_{-q}^{\alpha}) = (g\mu_B)^{-2} \chi_q^{\alpha} \tag{21}$$

was used. Hirahara et al.[5] have shown experimentally that the relation (20) actually holds in the case of MnP. They have measured the temperature dependence of the attenuation coefficient and the sound velocity in MnP in the critical temperature region. If the damping constant with $q \sim 0$ along the c-axis in MnP is written as Γ_0^c, this constant is related to the attenuation coefficient α_k and the velocity change Δv_l as[1),11),15]

$$\Gamma_0^c \propto -\frac{\omega_k^2 (\Delta v_l / v_l)}{v_l \alpha_k}. \tag{22}$$

Using this relation, Hirahara et al.[5] have obtained the result that Γ_0^c is proportional to $(T - T_C)^{1.1}$. The static susceptibility of MnP measured by Huber and Ridgley[16] is proportional to $(T - T_C)^{-1}$ in the critical region. Therefore, the relation (20) is consistent with the experimental result obtained by Hirahara et al.[5] It should be noted that Γ_q^{α} in our case is different from that for spin systems in insulators where spin damping occurs within the spin systems through spin diffusion. In the spin systems, when the exchange interaction is isotropic, $(K_q^{\alpha}(t), K_{-q}^{\alpha}(0))$ have anomaly at the transition temperature and the conventional theory cannot be used.

Solving the equation of motion (16), we obtain the two-spin relaxation function as follows: In the case that a quantity $|\Gamma_q'|$ defined by $|\Gamma_q{}^x-\Gamma_q{}^y|$ is much greater than the frequency of the collective motion of spins

$$\Omega_q=\sqrt{\Lambda_q{}^x\Lambda_q{}^y}\,, \tag{23}$$

we have

$$\left.\begin{aligned}&(S_q{}^\alpha(t), S_{-q}^\alpha(0))=(S_q{}^\alpha, S_{-q}^\alpha)\exp(-\Gamma_q{}^\alpha t),\\&(S_q{}^\alpha(t), S_{-q}^{\alpha'}(0))=0\,, \qquad (\alpha\neq\alpha')\end{aligned}\right\} \tag{24}$$

and in the case of $|\Gamma_q'|\ll\Omega_q$, we have

$$\left.\begin{aligned}&(S_q{}^x(t), S_{-q}^x(0))=(S_q{}^x, S_{-q}^x)\exp(-\Gamma_q t)\cos\Omega_q t\,,\\&(S_q{}^y(t), S_{-q}^y(0))=(S_q{}^y, S_{-q}^y)\exp(-\Gamma_q t)\cos\Omega_q t\,,\\&(S_q{}^z(t), S_{-q}^z(0))=(S_q{}^z, S_{-q}^z)\exp(-\Gamma_q{}^z t),\\&(S_q{}^x(t), S_{-q}^y(0))=-(S_q{}^y, S_{-q}^y)\sqrt{\frac{\Lambda_q{}^y}{\Lambda_q{}^x}}\exp(-\Gamma_q t)\sin\Omega_q t\,,\\&(S_q{}^y(t), S_{-q}^x(0))=(S_q{}^x, S_{-q}^x)\sqrt{\frac{\Lambda_q{}^x}{\Lambda_q{}^y}}\exp(-\Gamma_q t)\sin\Omega_q t\,,\\&(S_q{}^z(t), S_{-q}^x(0))=(S_q{}^x(t), S_{-q}^z(0))=(S_q{}^z(t), S_{-q}^y(0))\\&\qquad=(S_q{}^y(t), S_{-q}^z(0))=0\,,\end{aligned}\right\} \tag{25}$$

$$\Gamma_q=(\Gamma_q{}^x+\Gamma_q{}^y)/2\,. \tag{26}$$

Hereafter we assume that the wave number of applied sound is very small ($k\sim0$), as in usual cases. We assume also that $g_q{}^\alpha(\boldsymbol{k})$ is isotropic with respect to α and has a value $g_q(\boldsymbol{k})$. Inserting Eq. (24) into Eq. (15) and using the relation (21), we obtain α_k in the case $|\Gamma_q'|\gg\Omega_q$ as

$$\begin{aligned}\alpha_k=\frac{1}{\rho V v_l(g\mu_B)^4}\Bigg[&2|g_0(\boldsymbol{k})|^2(g\mu_B\langle S_0{}^z\rangle)^2\chi_0{}^z\frac{\Gamma_0{}^z}{(\Gamma_0{}^z)^2+\omega_k{}^2}\\&+\beta^{-1}\sum_q\sum_\alpha|g_q(\boldsymbol{k})|^2(\chi_q{}^\alpha)^2\frac{2\Gamma_q{}^\alpha}{(2\Gamma_q{}^\alpha)^2+\omega_k{}^2}\Bigg].\end{aligned} \tag{27}$$

In the case $|\Gamma_q'|\ll\Omega_q$, inserting Eq. (25) into Eq. (15), we have

$$\begin{aligned}\alpha_k=\frac{1}{\rho V v_l(g\mu_B)^4}\Bigg[&2|g_0(\boldsymbol{k})|^2(g\mu_B\langle S_0{}^z\rangle)^2\chi_0{}^z\frac{\Gamma_0{}^z}{(\Gamma_0{}^z)^2+\omega_k{}^2}\\&+\beta^{-1}\sum_q|g_q(\boldsymbol{k})|^2\left\{((\chi_q{}^x)^2+(\chi_q{}^y)^2)\frac{2\Gamma_q}{(2\Gamma_q)^2+\omega_k{}^2}\right.\\&\left.+(\chi_q{}^z)^2\frac{2\Gamma_q{}^z}{(2\Gamma_q{}^z)^2+\omega_k{}^2}\right\}\Bigg].\end{aligned} \tag{28}$$

In deriving Eq. (28), the terms proportional to $(\Lambda_q{}^y-\Lambda_q{}^x)/\Lambda_q{}^x$ were neglected, since $|(\Lambda_q{}^y-\Lambda_q{}^x)/\Lambda_q{}^x|$ is negligibly small in most of the magnetic materials. If

this term is included, α_k becomes dependent on the normal frequency of the collective motion of spins.

§ 3. Sound velocity

Using the relation (9), Kawasaki and Ikushima[15] have calculated the change in the sound velocity due to the spin-phonon interaction in the low frequency limit ($\omega_k \ll \Gamma_q^{\alpha}$). They expanded ($b_k$, b_k^*) in terms of the interaction Hamiltonian $\mathcal{H}_1$ and $\mathcal{H}_2$, and obtained the following expressions for the sound frequency shift

$$\Delta\omega_k = (\Delta\omega_k)_1 + (\Delta\omega_k)_2 , \tag{29}$$

$$(\Delta\omega_k)_1 = -(\beta/2\rho V\omega_k^0)\langle U_k^{(1)} U_{-k}^{(1)}\rangle , \tag{30}$$

$$(\Delta\omega_k)_2 = (1/\rho V\omega_k^0)\langle U_{kk}^{(2)}\rangle , \tag{31}$$

where ω_k^0 is the frequency in the absence of spin-phonon interaction. The frequency shift $(\Delta\omega_k)_1$ is the second order contribution from $\mathcal{H}_1$ and $(\Delta\omega_k)_2$ is the first order contribution from $\mathcal{H}_2$. In the case of the attenuation, the lowest order contribution arises only from the Hamiltonian $\mathcal{H}_1$. However, the contribution to the velocity change comes both from $\mathcal{H}_1$ and from $\mathcal{H}_2$. It is noticed that Eqs. (30) and (31) do not include the time-correlation. This is in contrast with the fact that the attenuation coefficient includes the time-correlation. The expressions (5) and (6) are respectively inserted into Eqs. (30) and (31), and the transformation (13) is made. In the same way as in the case of the attenuation, the four-spin correlation function appearing in the expression of velocity change is decoupled into a product of the two-spin correlation functions, and these two-spin correlation functions are replaced by the staggered susceptibilities, using the relation (21). Then, the calculated result for the velocitychange is given by

$$\Delta v_l = (\Delta v_l)_1 + (\Delta v_l)_2 , \tag{32}$$

$$\begin{aligned}(\Delta v_l)_1 &= (\Delta\omega_k)_1/k \\ &= -[\rho V v_l (g\mu_B)^4 k^2]^{-1}[2|g_0(\boldsymbol{k})|^2 (g\mu_B\langle S_0^z\rangle)^2\chi_0^z \\ &\quad + \beta^{-1}\sum_q\sum_{\alpha}|g_q(\boldsymbol{k})|^2(\chi_q^{\alpha})^2],\end{aligned} \tag{33}$$

$$\begin{aligned}(\Delta v_l)_2 &= (\Delta\omega_k)_2/k = -[2\rho V v_l (g\mu_B)^2 k^2]^{-1} \\ &\quad \times [h_0^z(\boldsymbol{k})(g\mu_B\langle S_0^z\rangle)^2 + \beta^{-1}\sum_q\sum_{\alpha} h_q^{\alpha}(\boldsymbol{k})\chi_q^{\alpha}],\end{aligned} \tag{34}$$

where $g_q(\boldsymbol{k})$ has been given by Eq. (12) and $h_q^{\alpha}(\boldsymbol{k})$ is defined by

$$h_q^{\alpha}(\boldsymbol{k}) = \sum_j e^{-i\boldsymbol{q}\cdot\boldsymbol{R}_{ji}}(e^{i\boldsymbol{k}\cdot\boldsymbol{R}_{ji}}-1)(e^{-i\boldsymbol{k}\cdot\boldsymbol{R}_{ji}}-1)\boldsymbol{e}_{\boldsymbol{k}}\boldsymbol{e}_{-\boldsymbol{k}} : \frac{\partial^2 J_{ij}^{\alpha}}{\partial\boldsymbol{R}_i\partial\boldsymbol{R}_j} . \tag{35}$$

The sign of $(\Delta v_l)_1$ is always negative and the spin-phonon interaction causes a dip, in the sound velocity versus temperature curve at the critical point. The

sign of $(\Delta v_l)_2$ depends on the sign of $h_q^{\alpha}(\boldsymbol{k})$. The velocity change $(\Delta v_l)_2$ has no singularity at the critical point, since this term originates from the rather short range correlation of spins inside the exchange interaction range. Therefore, as far as the critical anomaly is concerned, the contribution from $(\Delta v_l)_1$ dominates. In the presence of the magnetic field along the easy direction of magnetization of ferromagnets, the singularity in $(\Delta v_l)_1$ disappears, as will be shown in § 6. In this case, the contribution from $(\Delta v_l)_2$ cannot be neglected except $|g_q(\boldsymbol{k})|^2 \gg |h_q^{\alpha}(\boldsymbol{k})|$.

§ 4. Staggered susceptibilities in an external magnetic field

As seen from Eqs. (20), (27), (28), (33) and (34), the attenuation coefficient and the velocity change are expressed as functions of the staggered susceptibilities and the spin polarization. In this section we calculate the susceptibilities and the spin polarization, using the molecular field approximation and derive explicit formulae for the attenuation coefficient and the velocity change. Using the results calculated with the use of the formulae, we will discuss qualitatively the temperature and field dependence of these quantities in the next section. When the calculated attenuation coefficient of MnP is quantitatively compared with the experimental one in § 6, a certain correction for the staggered susceptibilities and the spin polarization is made.

When a static magnetic field H and a staggered magnetic field H_q^{α} are applied along the z- and α-axes, respectively, the Hamiltonian for a spin system is written as

$$\mathcal{H} = -\sum_{\alpha}\sum_{i,j} J_{ij}^{\alpha} S_i^{\alpha} S_j^{\alpha} + g\mu_B H \sum_i S_i^z + g\mu_B H_q^{\alpha} \sum_i S_i^{\alpha} e^{-i\boldsymbol{q}\cdot\boldsymbol{R}_i}. \tag{36}$$

From this Hamiltonian the molecular field Hamiltonian for the i-th spin is reduced to

$$\begin{aligned}\mathcal{H}_{mf}(i) = &\left(-2J_0^z \frac{\langle S_0^z\rangle}{\sqrt{N}} + g\mu_B H\right) S_i^z \\ &+ \left(-2J_q^{\alpha} \frac{\langle S_q^{\alpha}\rangle}{\sqrt{N}} + g\mu_B H_q^{\alpha}\right) S_i^{\alpha} e^{-i\boldsymbol{q}\cdot\boldsymbol{R}_i}.\end{aligned} \tag{37}$$

The value of $\langle S_0^z\rangle$ is determined by solving the self-consistency equation

$$\langle S_0^z\rangle/\sqrt{N} = S\sigma = -S\,\mathrm{B}_S(\beta S a), \tag{38}$$

$$a = -2J_0^z S\sigma + g\mu_B H, \tag{39}$$

where $\mathrm{B}_S(x)$ is the Brillouin function and σ is the spin polarization normalized by S. For the calculation of $\langle S_q^{\alpha}\rangle$, we consider the density matrix corresponding to the Hamiltonian (37) and expand it in terms of the last term of Eq. (37). Using this density matrix, we set up the self-consistency equation for $\langle S_q^{\alpha}\rangle$, and by solving this equation we obtain the staggered spin-polarization. If this stag-

gered spin-polarization is inserted into the relation

$$\chi_q{}^\alpha = -g\mu_B\langle S_q{}^\alpha\rangle/(\sqrt{N}H_q{}^\alpha), \tag{40}$$

the diagonal elements of the staggered susceptibility are obtained as

$$\chi_q{}^z = \frac{(g\mu_B)^2 S^2 B_S'(\beta Sa)}{\beta^{-1} - 2J_q{}^z S^2 B_S'(\beta Sa)}, \tag{41}$$

$$\chi_q{}^\alpha = \frac{(g\mu_B)^2 S B_S(\beta Sa)}{a - 2J_q{}^\alpha S B_S(\beta Sa)}, \qquad (\alpha = x, y) \tag{42}$$

where $B_S'(x) = (d/dx)B_S(x)$. In numerical calculations, we expand the Brillouin function and retain terms up to third order, since the magnitude of the normalized spin-polarization σ is less than 0.3 at the temperature and the field which we are concerned with. In this case, the susceptibilities (41) and (42) are reduced to

$$\chi_q{}^z = \frac{C(1-b)}{T - T_q{}^z(1-b)}, \tag{43}$$

$$\chi_q{}^\alpha = \frac{C(1-(b/3))}{T - T_q{}^\alpha(1-(b/3))}, \qquad (\alpha = x, y) \tag{44}$$

C being the Curie constant $(g\mu_B)^2 S(S+1)/3k_B$. In Eqs. (43) and (44), $T_q{}^\alpha$ is the ordering temperature for $S_q{}^\alpha$ and is given by

$$T_q{}^\alpha = \frac{2J_q{}^\alpha S(S+1)}{3k_B}, \tag{45}$$

and b is a constant defined by

$$b = \frac{1}{10}\beta^2[S^2 + (S+1)^2]a^2. \tag{46}$$

The maximum value of $T_q{}^\alpha$ with respect to q and α, $T_Q{}^\alpha$, corresponds to the ordering temperature at which the sinusoidal spin ordering with the wave number Q actually occurs. The ferromagnetic and antiferromagnetic spin-orderings are included in this scheme as special cases. The value $T_q{}^\alpha$ is expanded around $T_Q{}^\alpha$ as

$$T_q{}^\alpha = T_Q{}^\alpha - D_x(q_x - Q_x)^2 - D_y(q_y - Q_y)^2 - D_z(q_z - Q_z)^2, \tag{47}$$

$$D_\delta = -\frac{S(S+1)}{3k_B}\left.\frac{\partial^2 J_q}{\partial q_\delta{}^2}\right|_{q=Q}$$

$$= \frac{S(S+1)}{3k_B}\sum_i J_{ij}(R_i{}^\delta - R_j{}^\delta)^2 e^{iQ\cdot(R_i - R_j)}, \tag{48}$$

$$\delta = x, y, z,$$

where the α-dependence of J_{ij} was neglected in Eq. (48). The quantitics D_δ are more sensitive to the exchange interaction range than $J(Q)$, since it includes

$(R_i^\delta - R_j^\delta)^2$ in the sum.

In the case $|\Gamma_Q'| \gg \Omega_Q$, inserting Eqs. (20), (43), (44) and (47) into Eq. (27) and making the summation in Eq. (27) with respect to q, we obtain

$$\alpha_k = \frac{S^4(S+1)^2NT}{18\rho V v_l \mathrm{B}\, k_\mathrm{B}}\Bigg[4|g_0(k)|^2\left(\frac{\sigma(1-b)}{T-T_0^z(1-b)}\right)^2 + \frac{(1+(1/S))|g_Q(k)|^2 v_0 T}{96\pi\sqrt{D_xD_yD_z}}\Bigg\{\left(\frac{1-b}{T-T_Q^z(1-b)}\right)^{3/2} + \sum_{\alpha\neq z}\left(\frac{1-(b/3)}{T-T_Q^\alpha(1-(b/3))}\right)^{3/2}\Bigg\}\Bigg], \tag{49}$$

v_0 being the volume per spin, V/N. In deriving Eq. (49), ω_k in the denominator of Eq. (27) was neglected. In the same way, we obtain α_k for the case $|\Gamma_Q'| \ll \Omega_Q$ as

$$\alpha_k = \frac{S^4(S+1)^2NT}{18\rho V v_l \mathrm{B}\, k_\mathrm{B}}\Bigg[4|g_0(k)|^2\left(\frac{\sigma(1-b)}{T-T_0^z(1-b)}\right)^2 + \frac{(1+(1/S))|g_Q(k)|^2 v_0 T}{96\pi\sqrt{D_xD_yD_z}}\Bigg\{\left(\frac{1-b}{T-T_Q^z(1-b)}\right)^{3/2} + \frac{16}{(1-(b/3))^{1/2}(T_Q^x - T_Q^y)^2} \times\left(\frac{\{[T-T_Q^x(1-(b/3))]^{1/2} - [T-((T_Q^x+T_Q^y)/2)(1-(b/3))]^{1/2}\}^2}{[T-T_Q^x(1-(b/3))]^{1/2}} + \frac{\{[T-T_Q^y(1-(b/3))]^{1/2} - [T-((T_Q^x+T_Q^y)/2)(1-(b/3))]^{1/2}\}^2}{[T-T_Q^y(1-(b/3))]^{1/2}}\right)\Bigg\}\Bigg]. \tag{50}$$

If Eqs. (43), (44) and (47) are inserted into Eqs. (33) and (34), the velocity change due to the spin-phonon interaction is calculated as

$$(\Delta v_l)_1 = -\frac{NS^3(S+1)}{6\rho V v_l k^2 k_\mathrm{B}}\Bigg[4|g_0(k)|^2\sigma^2\left(\frac{1-b}{T-T_0^z(1-b)}\right) + \frac{(1+(1/S))|g_Q(k)|^2 v_0 T}{12\pi\sqrt{D_xD_yD_z}}\Bigg\{\left(\frac{1-b}{T-T_Q^z(1-b)}\right)^{1/2} + \sum_{\alpha\neq z}\left(\frac{1-(b/3)}{T-T_Q^\alpha(1-(b/3))}\right)^{1/2}\Bigg\}\Bigg], \tag{51}$$

$$(\Delta v_l)_2 = -\frac{NS^2}{2\rho V v_l k^2 k_\mathrm{B}}\Bigg[h_0^z(k)\sigma^2 - \frac{(1+(1/S))v_0 T}{12\pi\sqrt{D_xD_yD_z}}\Bigg\{h_Q^z(k) \times\left(\frac{T-T_Q^z(1-b)}{1-b}\right)^{1/2} + \sum_{\alpha\neq z} h_Q^\alpha(k)\left(\frac{T-T_Q^\alpha(1-(b/3))}{1-(b/3)}\right)^{1/2}\Bigg\}\Bigg]. \tag{52}$$

In Eq. (52), a constant term independent of temperature and magnetic field was neglected. When both magnetic field and anisotropy are in absence in ferromagnets, the last terms in the curly brackets of Eqs. (49), (50) and (51), which originate from the fluctuations of the x- and y-components of spin, diverge below T_Q. In this case, the correlation length of the x- and y-components of the spin correlation function becomes infinite. Therefore, we cannot use the decoupling approximation which was employed in deriving Eqs. (15) and (33). However, when anisotropy or magnetic field exists, the correlation length becomes finite,[17] and therefore the formulae may be used even below the transition temperature.

§ 5. Weak magnetic field limit

Let us consider the temperature dependence of the sound attenuation in a constant magnetic field. When temperature is very near the phase transition temperature, the sound attenuation always decreases in an applied magnetic field because of the suppression of spin fluctuations by the field. When the temperature increases, this negative contribution decreases rapidly, and is completely compensated by the positive contribution from the first term in Eq. (14) at a certain temperature. When the temperature increases furthermore, the positive contribution from the spin polarization overcomes the negative contribution from the suppression of spin-fluctuations, and the attenuation begins to increase. As shown below, the compensation temperature is closely related to the nature of the exchange interaction in magnetic materials. In ferromagnets with the long range exchange interaction the compensation temperature becomes extremely near to the transition temperature, whereas in antiferromagnets with the short range interaction the temperature becomes very high away from the transition temperature.

The temperature range in which the critical attenuation is usually measured is from 10^{-4} to 10^{-1} of $(T-T_Q)/T_Q$. If the temperature is higher than the compensation temperature, the attenuation is observed to increase in the presence of magnetic field. On the other hand, if the temperature is lower than the compensation temperature, the attenuation is observed to decrease in the field. To examine the relation between the exchange interaction and the compensation temperature, we consider the attenuation coefficient in the weak field limit. We assume that the exchange interaction and the spin damping constant are isotropic. In this case the field-dependent part of the attenuation coefficient is obtained from Eq. (50) as

$$\Delta\alpha_{\boldsymbol{k}}(H)=\frac{NS^3(S+1)^3(g\mu_{\mathrm{B}}H)^2T}{54\rho V v_l \mathrm{B}\, k_{\mathrm{B}}{}^3}\left[\frac{4|g_0(\boldsymbol{k})|^2S(S+1)}{3(T-T_0)^4}\right.$$
$$\left.-\frac{|g_Q(\boldsymbol{k})|^2v_0[S^2+(S+1)^2]T^2}{128\pi\sqrt{D_xD_yD_z}(T-T_0)^2(T-T_Q)^{5/2}}\right]. \tag{53}$$

The first and second terms in the bracket of Eq. (53) come from the first and

second terms in Eq. (50), respectively. We notice that the field dependent part of α_z starts from a term proportional to H^2. The compensation temperature is determined by the condition that the magnitude of the first term in Eq. (53) becomes equal to that of the second term. We write the compensation temperature measured from T_Q as ΔT. In the case that T_Q and T_Q-T_0 are larger than ΔT, ΔT is calculated as

$$(\Delta T)^{5/2}=\frac{3v_0[S^2+(S+1)^2]|g_Q(\boldsymbol{k})|^2T_Q^2(T_Q-T_0)^2}{512\pi S(S+1)\sqrt{D_xD_yD_z}|g_0(\boldsymbol{k})|^2}. \tag{54}$$

If we indicate the phase transition temperature by T_C and the paramagnetic Curie temperature by Θ, T_Q-T_0 is equal to $T_C-\Theta$, and thus ΔT is proportional to $(T_C-\Theta)^{4/5}$. The value of ΔT is inversely proportional to $(D_xD_yD_z)^{1/5}$, which is sensitive to the exchange interaction range, as mentioned in § 4. In Dy and Ho which have a spin-spiral structure at high temperatures, $T_C-\Theta$ is considerably smaller than the Néel temperature. Furthermore, the exchange interaction in these metals is of the long range, since the interaction comes mainly from the indirect coupling through conduction electrons. Consequently, we expect that ΔT is very small in the rare earth metals.

In ferromagnets in which T_C-T_0 vanishes, ΔT is calculated from Eq. (53) as

$$(\Delta T)^{1/2}=\frac{3v_0[S^2+(S+1)^2]T_0^2}{512\pi S(S+1)\sqrt{D_xD_yD_z}}. \tag{55}$$

The value of ΔT in this case is inversely proportional to $D_xD_yD_z$, and thus ΔT is extremely sensitive to the exchange interaction range. Since in ferromagnets such as Gd and MnP the exchange interaction is of the long range, the compensation temperature is expected to be very near the phase transition temperatures. This fact is consistent with the result that the sound attenuation of these crystals has been observed to increase in an external magnetic field, except extremely near the transition temperatures.[18),5)]

Manganese fluoride is a typical antiferromagnet with the Néel temperature of 67 K and the paramagnetic Curie temperature of -82 K,[19)] and, thus, $T_C-\Theta$ of this crystal is 149 K, which is very large compared with the Néel temperature. Furthermore, the exchange interaction of this crystal is of the short range, since the second nearest neighbor interaction (the interaction between the corner and body centered sites in the rutile structure) dominates other interactions. The value of $v_0T_C^{3/2}(D_xD_yD_z)^{-1/2}$ of MnF_2 is estimated as 45. If this value, the Néel and paramagnetic Curie temperatures, and the manganese spin value of 5/2 are inserted into Eq. (53), and if the relation $|g_0(\boldsymbol{k})|^2=|g_Q(\boldsymbol{k})|^2$ is assumed, we find that Eq. (53) does not vanish except at infinite temperature. In other words, the compensation temperature goes to infinity and the sign of $\Delta\alpha_k(H)$ is always negative in the paramagnetic phase. This is consistent with the fact that the

sound attenuation of this crystal has been observed to decrease by an application of magnetic field in the temperature range of the paramagnetic phase.[2] One might suspect that the relation for spin damping constant (20) may not be correct for MnF_2 as mentioned in § 2, because this crystal is an insulator. However, our essential conclusion that $\Delta\alpha_k(H)$ is always negative may hold, even if a damping constant calculated from other mechanisms proper for MnF_2 is used.

§ 6. Manganese phosphide

a) Attenuation

Manganese phosphide has an orthorhombic crystal structure slightly distorted from the hexagonal structure of the NiAs-type. The spin structure as a function of temperature and the magnetic field along the b axis is shown in Fig. 1.[20] As seen in the figure, in the absence of magnetic field this crystal is ferromagnetic between 47 K and 290.5 K, and paramagnetic above 290.5 K. The transition temperature between these phases decreases with increasing magnetic field. At low temperatures, spin-spiral and spin-fan structure phases appear, respectively, in low and high magnetic fields. In the ferromagnetic phase, the easy, intermediate and hard axes of magnetization are the c, b and a axes, respectively.

Hirahara et al.[5] have obtained the following experimental results of the ultrasonic attenuation in MnP. (1) When a magnetic field is applied along the c axis, the height of the attenuation peak at the Curie temperature decreases abruptly and the width of the peak increases, as shown in Fig. 2. At a fixed temperature above the compensation temperature mentioned in § 5, the attenuation coefficient starts to increase proportionally to the square of the magnetic

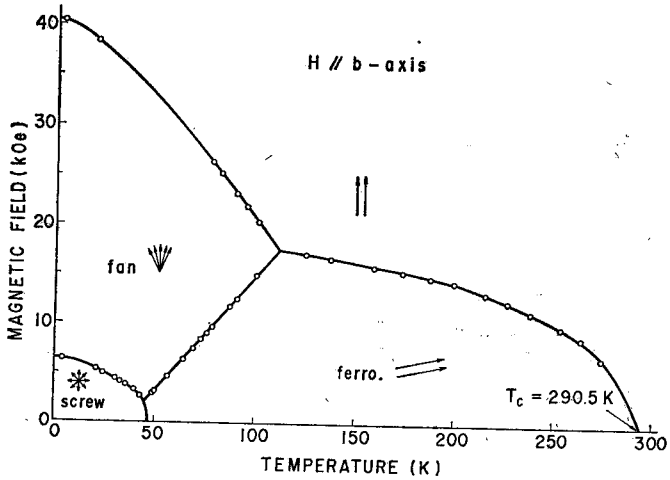

Fig. 1. Spin structure diagram as a function of temperature and the magnetic field along the b axis.

field, and attains a maximum at a certain magnetic field. With further increasing of the field, the attenuation decreases. The attenuation maximum shifts toward the higher magnetic field as temperature increases. (2) When a magnetic field is applied along the *a*- and *b*-axes, the temperature of the second order phase transition between the ferromagnetic and paramagnetic phases shifts toward the lower temperature. In addition to the attenuation peak at the shifted transition temperature, Hirahara et al.[5] have found a new broad peak above the Curie temperature.

To explain the experimental results mentioned above, we calculated the attenuation coefficient of MnP around the Curie temperature. When a magnetic field is applied along the *c* axis, Eqs. (49) and (50) are, respectively, reduced to

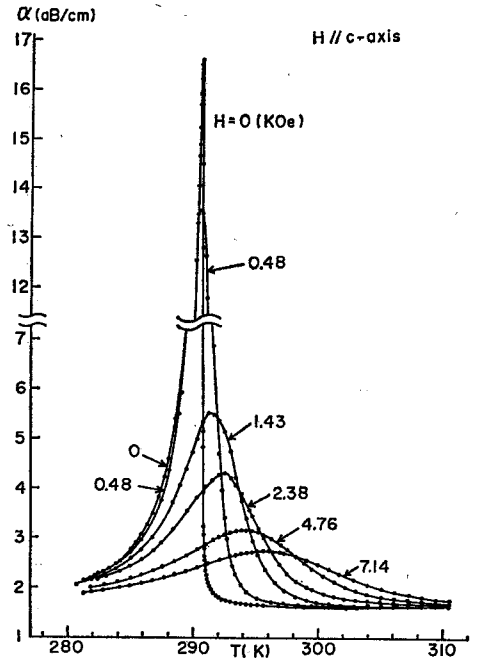

Fig. 2. Experimental results of the ultrasonic attenuation in MnP obtained by Hirahara et al.[5] Magnetic fields were parallel to the *c* axis. The longitudinal sound wave of 90 MHz propagated along the *b* axis.

$$\alpha_k^c = F_k t\left[\left(\frac{\sigma(1-b)}{t-1+b}\right)^2 + Gt\left\{\left(\frac{1-b}{t-1+b}\right)^{3/2} + \left(\frac{1-(b/3)}{t-(1-\kappa_a)(1-(b/3))}\right)^{3/2} + \left(\frac{1-(b/3)}{t-(1-\kappa_b)(1-(b/3))}\right)^{3/2}\right\}\right], \tag{56}$$

$$\alpha_k^c = F_k t\left[\left(\frac{\sigma(1-b)}{t-1+b}\right)^2 + Gt\left\{\left(\frac{1-b}{t-1+b}\right)^{3/2} + \frac{16}{(1-(b/3))^{1/2}(\kappa_a-\kappa_b)^2} \times \left(\frac{\{[t-(1-\kappa_a)(1-(b/3))]^{1/2} - [t-(1-((\kappa_a+\kappa_b)/2))(1-(b/3))]^{1/2}\}^2}{[t-(1-\kappa_a)(1-(b/3))]^{1/2}} + \frac{\{[t-(1-\kappa_b)(1-(b/3))]^{1/2} - [t-(1-((\kappa_a+\kappa_b)/2))(1-(b/3))]^{1/2}\}^2}{[t-(1-\kappa_b)(1-(b/3))]^{1/2}}\right)\right\}\right], \tag{57}$$

where

$$F_k = \frac{2S^4(S+1)^2|g_0(\boldsymbol{k})|^2 N}{9\rho V v_l \mathrm{B}\, k_B T_C}, \tag{58}$$

$$G=\frac{(1+(1/S))v_0T_C^{3/2}}{384\pi\sqrt{D_xD_yD_z}}, \tag{59}$$

$$t=T/T_C, \tag{60}$$

$$\kappa_a=\frac{J_0^c-J_0^a}{J_0^c}=\frac{2}{3}S(S+1)\frac{J_0^c-J_0^a}{k_BT_C}, \tag{61}$$

$$\kappa_b=\frac{J_0^c-J_0^b}{J_0^c}=\frac{2}{3}S(S+1)\frac{J_0^c-J_0^b}{k_BT_C}. \tag{62}$$

The value of F_k and G are considered as adjustable parameters and determined by comparison with experiment. The anisotropy parameters κ_a and κ_b are experimentally determined as follows: The anisotropy energy of MnP is expressed as

$$F_{\text{anis}}=K_a\alpha_a^2+K_b\alpha_b^2, \tag{63}$$

where α_a and α_b are, respectively, the direction cosines of magnetization with respect to the a- and b-axes. From torque measurements, Huber and Ridgley[16] have obtained the anisotropy constants K_a and K_b. Hiyamizu and Nagamiya[21] have extrapolated their values from 47 K to absolute zero and obtained

$$K_a=2.86\text{ K} \quad \text{and} \quad K_b=0.89\text{ K per atom}. \tag{64}$$

Since these quantities are related to $J_0^c-J_0^a$ and $J_0^c-J_0^b$ by the relations $S^2(J_0^c-J_0^a)=K_a$ and $S^2(J_0^c-J_0^b)=K_b$, κ_a and κ_b are calculated from Eqs. (61) and (62) as

$$\kappa_a=0.0131 \quad \text{and} \quad \kappa_b=0.0039. \tag{65}$$

We assumed that S is equal to one.

First we calculate the attenuation coefficient when temperature is fixed and magnetic field is changed.*) The staggered susceptibilities for small q make dominant contributions to the attenuation in ferromagnets such as MnP. For calculating the susceptibilities, we used the molecular field approximation. However, the uniform susceptibility calculated by this approximation is not in agreement quantitatively with the experimental values obtained by Huber and Ridgley[16] in the critical temperature region. Therefore, for making a correction to this discrepancy, an effective temperature T' is introduced, which is given as a function of T by the equation

$$\chi^c_{\text{calculated}}(T')=\chi^c_{\text{observed}}(T). \tag{66}$$

Between the temperature of 290.5 K and 330.5 K, the susceptibility obtained by Huber and Ridgley[16] is expressed as

*) Since Eq. (39) includes the g-value, the g-value for MnP is needed for calculating the spin polarization and the susceptibilities for a given strength of magnetic field. We used the g-value of 2.05 which has been obtained by Kunii and Hirahara using ferromagnetic resonance technique (private communication).

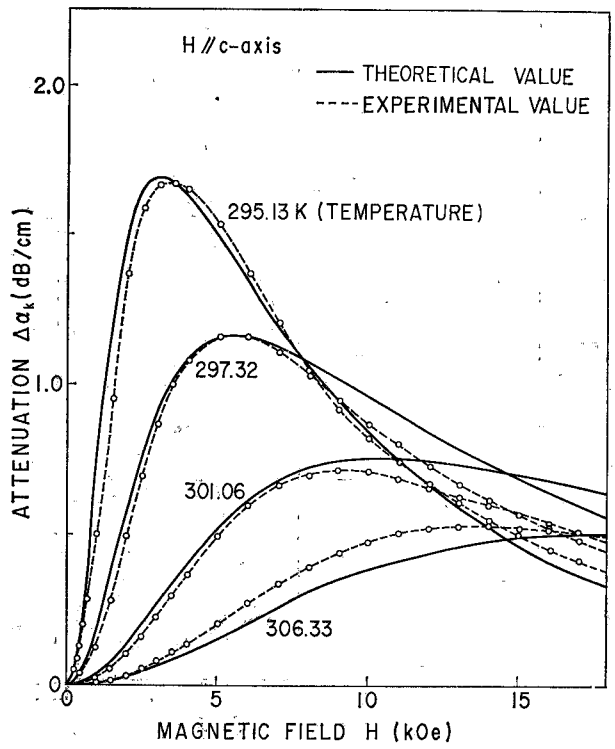

Fig. 3. Magnetic field dependence of the attenuation coefficient of MnP measured from that in zero-field; $\Delta\alpha_k(H)=\alpha_k(H)-\alpha_k(0)$. The magnetic field was applied along the c axis. Solid lines represent the calculated values and white circles show the experimental values obtained by Hirahara et al.[5] They used the longitudinal sound wave of 90 MHz propagating along the b axis. The two parameters F_k and G appearing in Eqs. (56) and (57) were determined from the following experimental values for 297.32 K: (i) 6kOe at which the attenuation attains a maximum and (ii) the magnitude of the attenuation coefficient at 6kOe.

$$\chi^{c}_{\text{observed}}(T)=\frac{C'}{T-T_c}, \tag{67}$$

with $C'=C/0.5905$. Therefore, Eq. (66) gives the effective temperature as

$$T'=T_c+\frac{C}{C'}(T-T_c). \tag{68}$$

Since in MnP the condition $|\Gamma_q'|\ll\Omega_q$ is expected to be fulfilled in almost the whole temperature and field range of interest, Eq. (57) is used for the calculation of α_k. However, the difference between the values of α_k which are calculated from Eqs. (56) and (57) is very small in the case of MnP. For determining the adjustable parameter G, we calculated α_k/F_k as a function of the magnetic field and the parameter G, fixing temperature at 297.32 K. The calculated α_k/F_k

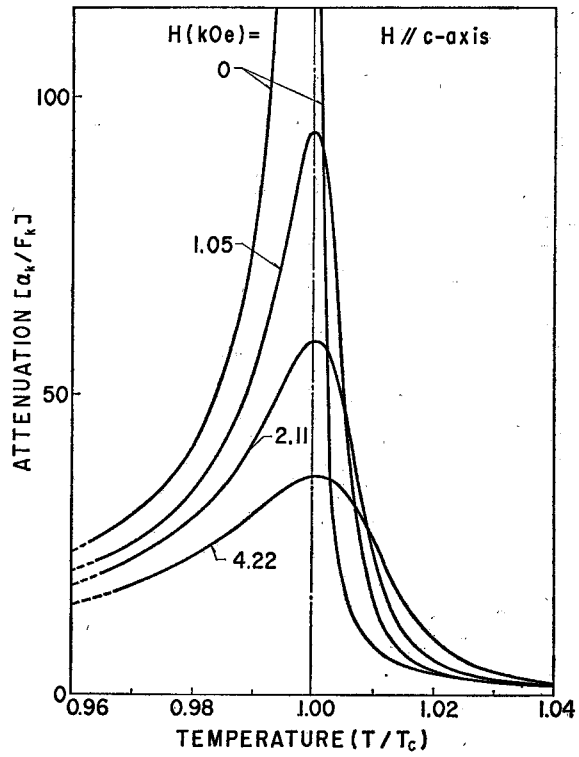

Fig. 4. Temperature dependence of the attenuation coefficient of MnP in magnetic fields applied along the c axis. Dotted lines show the coefficient in the temperature range where the spin polarization exceeds 0.3.

has a maximum at a certain magnetic field whose strength depends on the magnitude of G and coincides with the experimental value of 6 kOe at the value of G of 1/240. The value of F_k is determined from the magnitude of the attenuation at the maximum point. Using these values of F_k and G, we obtain the field dependence of the attenuation coefficient for various temperatures, and show the results in Fig. 3. As seen from the figure, agreement between theory and experiment is satisfactory. The temperature dependence of the attenuation for several magnetic fields is obtained using the same parameter of G and shown in Fig. 4. In Fig. 4, we did not use the effective temperature, since we could not determine the effective temperature below the Curie temperature. Therefore, the theoretical result may qualitatively be compared with the experimental one shown in Fig. 2. From Fig. 4 we see that the divergence in the attenuation at the Curie temperature disappears, corresponding to disappearance of the second order phase transition in ferromagnets in a magnetic field along the easy direction of magnetization. This divergence originates from the divergence of the component of spin fluctuation with $q=0$ at the Curie temperature. Since this fluctuation is

suppressed by a uniform magnetic field, the divergence in the attenuation disappears. On the other hand, in antiferromagnets and spin-spiral structures, the divergence in the attenuation at phase transition temperatures originates from the spin fluctuation with a finite value of q. Since the divergence in this spin fluctuation is not suppressed by the uniform magnetic field, the divergence in the attenuation does not disappear in these cases. Using this attenuation peak, Shapira et al.[22)~25)] have made intensive studies to determine phase boundaries in the magnetic phase diagrams in many antiferromagnets. As will be discussed in the following, the divergence in the attenuation does not disappear also when a magnetic field is applied along the hard axes in ferromagnets.

When a magnetic field is applied along the b axis, Eqs. (49) and (50) are respectively reduced to

$$\alpha_k{}^b = F_k t\left[\left(\frac{\sigma(1-b)}{t-(1-\kappa_b)(1-b)}\right)^2 + Gt\left(\frac{1-b}{t-(1-\kappa_b)(1-b)}\right)^{3/2} + Gt\left(\frac{1-(b/3)}{t-(1-\kappa_a)(1-(b/3))}\right)^{3/2} + Gt\left(\frac{1-(b/3)}{t-1+(b/3)}\right)^{3/2}\right], \tag{69}$$

$$\begin{aligned}\alpha_k{}^b = F_k t\Bigg[&\left(\frac{\sigma(1-b)}{t-(1-\kappa_b)(1-b)}\right)^2 + Gt\left(\frac{1-b}{t-(1-\kappa_b)(1-b)}\right)^{3/2} \\ &+ \frac{16Gt}{(1-(b/3))^{1/2}\kappa_a{}^2} \\ &\times \frac{\{[t-(1-\kappa_a)(1-(b/3))]^{1/2} - [t-(1-(\kappa_a/2))(1-(b/3))]^{1/2}\}^2}{[t-(1-\kappa_a)(1-(b/3))]^{1/2}} \\ &+ \frac{16Gt}{(1-(b/3))^{1/2}\kappa_a{}^2} \frac{\{[t-1+(b/3)]^{1/2} - [t-(1-(\kappa_a/2))(1-(b/3))]^{1/2}\}^2}{[t-1+(b/3)]^{1/2}}\Bigg].\end{aligned} \tag{70}$$

The spin component along the b axis is induced by an external magnetic field in all the phases in the magnetic phase diagram shown in Fig. 1. On the other hand, the spin component along the c axis aligns cooperatively below the boundary critical temperature $T_C{}^*$ between the paramagnetic and ferromagnetic phases. From Eq. (44) the susceptibility along the c axis is reduced to

$$\chi_0{}^c = \frac{C(1-(b/3))}{T-T_C(1-(b/3))}. \tag{71}$$

The denominator of Eq. (71) vanishes at $T_C{}^*$ and $\chi_0{}^c$ diverges. Therefore, $T_C{}^*$ is determined by the equation

$$T_C{}^* - T_C\left(1-\frac{b}{3}\right) = 0, \tag{72}$$

where b is a function of temperature and magnetic field, and calculated from Eqs. (38), (39), and (46). The temperature $T_C{}^*$ calculated from Eq. (72) de-

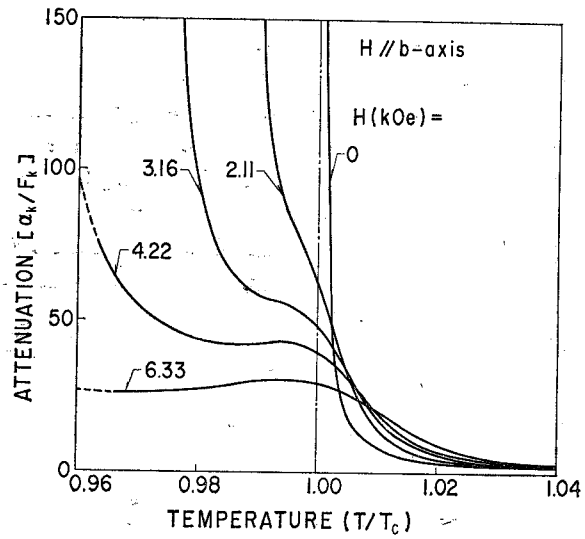

Fig. 5. Temperature dependence of the attenuation coefficient of MnP in magnetic fields applied along the b axis. The dotted lines show the coefficient in the temperature range where the spin polarization exceeds 0.3.

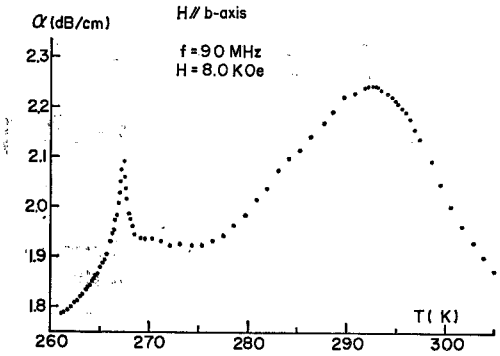

Fig. 6. Experimental values of the ultrasonic attenuation in MnP obtained by Hirahara et al.[5] Both the magnetic field of 8.0kOe and the propagation of the longitudinal sound wave of 90 MHz are parallel to the b axis.

creases as the magnetic field increases. From Eq. (72), the denominators of the last terms of Eqs. (69) and (70) vanish at T_C^*. Therefore, the divergence in the attenuation occurs at T_C^*.

The attenuation coefficient was calculated from Eqs. (69) and (70) using the parameter G which has been determined above, and the result is shown in Fig. 5. As seen in this figure, T_C^* at which the attenuation coefficient diverges shifts toward the lower temperature as the magnetic field increases. When the magnetic field is weak, the attenuation coefficient *vs.* temperature curve has a shoulder. This shoulder grows into a broad peak around $T \sim T_C$ when the magnetic field is strong enough and T_C^* lowers down far away from T_C. This broad peak originates from the first and second terms in the brackets in Eqs. (69) and (70). This means that the spin fluctuation along the b axis has its maximum at $T \sim T_C$, even when T_C^* shifts toward a lower temperature in the presence of magnetic field. As discussed above, the fluctuations of the spin components along the c- and b-axes causes anomalies in the attenuation respectively at T_C^* and $T \sim T_C$ in the presence of magnetic field. Hirahara et al.[5] have made ultrasonic measurements on MnP in the magnetic fields, using the frequency of 90 MHz, and actually observed sharp peaks at T_C^* and broad peaks around T_C. As an example the temperature dependence of the attenuation coefficient for 8.0 kOe is shown in Fig. 6.

When a magnetic field is applied along the a axis, the attenuation coefficient is calculated by the expressions in which the suffixies a and b in Eqs. (69) and (70) are interchanged. The characteristics of the critical attenuation in this case is similar to that in the previous case.

The ultrasonic energy is dissipated by conduction electrons through the electron-phonon interaction. We estimate the contribution to the attenuation coefficient of MnP from this mechanism. According to Pippard,[26] when the wave length of longitudinal sound is longer than the mean free path of conduction electron, the attenuation coefficient is expressed as

$$\alpha_k = \frac{4\hbar^2 k_F^2 \sigma \omega_k^2}{15 e^2 \rho v_l^3}, \qquad (73)$$

where k_F is the wave number of conduction electron at the Fermi surface, σ the conductivity, e the electronic charge, ρ the crystal density, v_l the velocity of longitudinal sound. The experimental value of σ of MnP at T_C is $4.0\times10^3(\Omega\cdot\mathrm{cm})^{-1}$,[27] the value of ρ is $5.8\mathrm{gr}\cdot\mathrm{cm}^{-3}$, and the value of v_l is $7.1\times10^5\,\mathrm{cm}\cdot\mathrm{sec}^{-1}$. Since we can not find the value of k_F for MnP, we use the value of $1.4\times10^8\,\mathrm{cm}^{-1}$ for metallic copper,[28] which may be larger than the value of k_F for MnP. Inserting these values into Eq. (73), α_k for the sound frequency of 100 MHz is estimated to be 1.2×10^{-4} dB/cm, which is negligibly small compared with the contribution from the spin-phonon interaction. In a magnetic field, the spins are polarized and the scattering of conduction electrons by the spin fluctuations is diminished. This causes an increase of the conductivity[29),30] (or negative resistance) and thus an increase of the attenuation. However, the magnitude of this increase is also negligibly small.

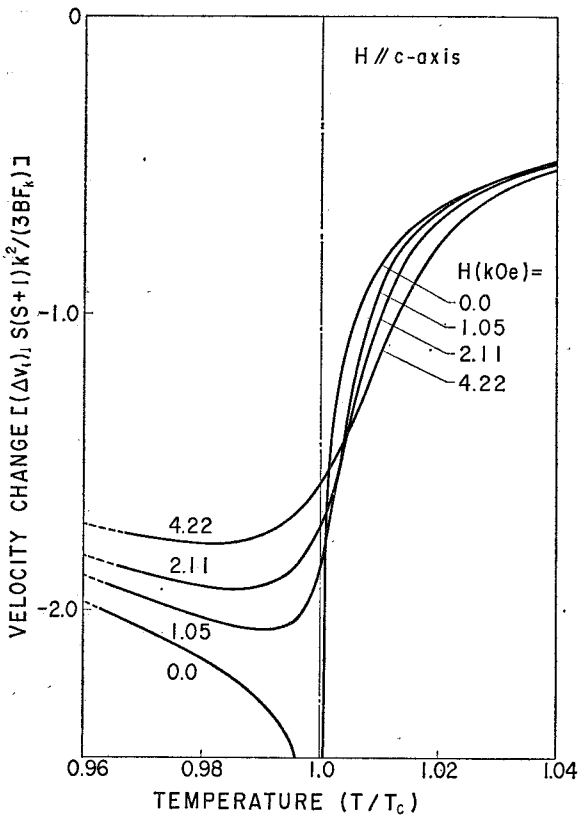

Fig. 7. Temperature dependence of the velocity change in MnP in magnetic fields applied along the c axis. Dotted lines show the velocity change in the temperature range where the spin polarization exceeds 0.3.

b) Velocity change

The velocity change due to the spin-phonon interaction is contributed from

the two terms $(\Delta v_l)_1$ and $(\Delta v_l)_2$ which have been given by Eqs. (51) and (52), respectively. The velocity change $(\Delta v_l)_1$ includes divergent terms at transition points, whereas $(\Delta v_l)_2$ does not have any anomalous term. Therefore, as far as we are concerned with the critical velocity change at the phase transition points in the absence of magnetic field, we may neglect the contribution from $(\Delta v_l)_2$. However, in finite magnetic fields, the divergence in the velocity change of $(\Delta v_l)_1$ disappears, similarly to the case of the attenuation, and thus the contribution from $(\Delta v_l)_2$ is not neglected if $|g_q(\boldsymbol{k})|^2 \lesssim |h_q{}^a(\boldsymbol{k})|$. Unfortunately, we do not have any information about $h_q{}^a(\boldsymbol{k})$ of MnP. Therefore, we calculate only the contribution from $(\Delta v_l)_1$. When a magnetic field is applied along the c axis, Eq. (51) is reduced to

$$(\Delta v_l)_1 = -F_k \frac{3B}{S(S+1)k^2}\left[\sigma^2\left(\frac{1-b}{t-1+b}\right)+8Gt\left\{\left(\frac{1-b}{t-1+b}\right)^{1/2}\right.\right.$$
$$\left.\left.+\left(\frac{1-(b/3)}{t-(1-\kappa_a)(1-(b/3))}\right)^{1/2}+\left(\frac{1-(b/3)}{t-(1-\kappa_b)(1-(b/3))}\right)^{1/2}\right\}\right]. \quad (74)$$

Substituting the value of G obtained in the previous subsection into Eq. (74), we calculated the velocity change and show the result in Fig. 7. As seen in this figure, the divergence in the velocity change at the Curie temperature disappears in magnetic fields. In the fields, the velocity decreases above the Curie temperature and increases below the Curie temperature. This behavior has been observed by Hirahara et al.[5] in MnP. The same behavior of the velocity change in magnetic fields has also been reported in Gd, Tb, Dy and Ho by Lüthi et al.[1]

§ 7. Dysprosium

The dysprosium metal has a spin-spiral structure between the temperatures of 181 K[3] and 85 K. Its spiral axis is along the c axis and spins are in the c plane of the hexagonal close packed crystal structure. Lee and Levy[3] have made the ultrasonic attenuation measurements on the dysprosium metal under a magnetic field applied in the c plane and found that the attenuation is strongly enhanced by an application of the magnetic field. The paramagnetic Curie temperature when a magnetic field is applied in the c plane is 169 K[31] which is only 12 K below the Néel temperature. The exchange interaction is of the long range, since the interaction comes mainly from the indirect coupling through conduction electrons. Therefore, as discussed in § 5, the compensation temperature is expected to be very near the Néel temperature. Actually, Lee and Levy[3] have observed an increase of the attenuation in the entire temperature range of the paramagnetic phase.

We calculated the attenuation coefficient in the magnetic field of 5 kOe in the c plane, using Eq. (50). The anisotropy of $T_Q{}^a$ which is uniaxial in this crystal was determined from the anisotropy of the paramagnetic Curie tempera-

ture. In Fig. 8, we show the calculated result of the attenuation coefficient for the six values of A which is defined by

$$A=\frac{(1+(1/S))v_0T_Q^{5/2}|g_Q(\boldsymbol{k})|^2}{384\pi\sqrt{D_xD_yD_z}|g_0(\boldsymbol{k})|^2}\,. \tag{75}$$

In the magnetic field of 5 kOe, the Néel temperature shifts to the lower temperature by 1 K. In Fig. 8, the attenuation coefficient in the absence of magnetic field, which was calculated by using Eq. (49), is shown by a dotted line. The factor $|g_Q(\boldsymbol{k})|^2/|g_0(\boldsymbol{k})|^2$ in Eq. (75) may be of the order of unity. The parameter A is a measure of the range of exchange interaction. As the range increases, $D_xD_yD_z$ in Eq. (75) increases as mentioned in § 4, and thus A decreases. If the result in Fig. 8 is compared with the experimental result of Lee and Levy,[3] the value of A is seen to be smaller than 1/300. This value is compared with 1/19 of A in MnF_2 which is a typical antiferromagnet having the short range exchange interaction.

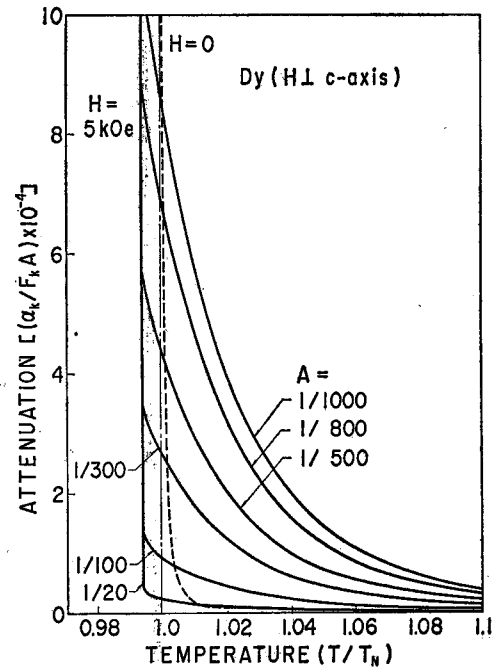

Fig. 8. Temperature dependence of the attenuation coefficient of Dy in the magnetic field of 5kOe for several values of the parameter A. This parameter is defined in Eq. (75) and is a measure of the exchange interaction range.

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Appendix

Another derivation of the attenuation coefficient—the first term in Eq. (14)

In the text, we have discussed that the cross term of the spin polarization and the two-spin relaxation function in the expression of α_k plays an important

role in the field dependence of α_k. To elucidate the physical origin of this term, we show another derivation in this Appendix.

The modulation of the exchange interaction due to a sound wave is written as

$$\mathcal{H}=-\sum_{i,j}\sum_{\alpha}\left(\boldsymbol{u}_i\cdot\frac{\partial J_{ij}^{\alpha}}{\partial \boldsymbol{R}_i}+\boldsymbol{u}_j\cdot\frac{\partial J_{ij}^{\alpha}}{\partial \boldsymbol{R}_j}\right)S_i^{\alpha}S_j^{\alpha}, \tag{A·1}$$

where $\boldsymbol{u}_i$ is the displacement of i-th atom by the sound propagation and is expressed as

$$\boldsymbol{u}_i=\boldsymbol{e}_k u_0\exp(i(\boldsymbol{k}\cdot\boldsymbol{R}_i-\omega_k t)). \tag{A·2}$$

We assume the spins polarized in the direction of z axis. If the spin component S_i^z or S_j^z in Eq. (A·1) is replaced by the static spin polarization, the z component of the Hamiltonian (A·1) is reduced to

$$\mathcal{H}'=2g_0^z(\boldsymbol{k})u_0\langle S_0^z\rangle S_k^z\exp(-i\omega_k t), \tag{A·3}$$

where $g_0^z(\boldsymbol{k})$ is defined by Eq. (12), and S_0^z and S_k^z are the Fourier amplitudes of spin. The remaining terms in Eq. (A·1) contribute to the second term in Eq. (14). The Hamiltonian (A·3) is equivalent to the Hamiltonian of S_k^z in the oscillating magnetic field $H_k\exp(-i\omega_k t)$:

$$\mathcal{H}'=g\mu_B H_k\exp(-i\omega_k t)S_k^z, \tag{A·4}$$

with

$$H_k=2g_0^z(\boldsymbol{k})u_0\langle S_0^z\rangle/g\mu_B. \tag{A·5}$$

The energy dissipation of the spin system or the energy dissipation from the sound wave is expressed as

$$P=\frac{\omega_k}{2}\chi_k''(\omega_k)H_kH_k^*, \tag{A·6}$$

$\chi_k''(\omega_k)$ being the imaginary part of the dynamical susceptibility. The attenuation coefficient for the sound amplitude is given by

$$\alpha_k=\frac{P}{2Ev_l}, \tag{A·7}$$

where E is the energy of the sound wave in a crystal and is expressed as $V\rho\omega_k^2u_0^2/2$. The factor 2 in the dinominator of Eq. (A·7) comes from the fact that the sound energy is proportional to the square of the amplitude of sound wave. Inserting Eq. (A·6) into Eq. (A·7), α_k is obtained as

$$\alpha_k=\frac{2|g_0^z(\boldsymbol{k})|^2\langle S_0^z\rangle^2\chi_k''(\omega_k)}{\rho V v_l\omega_k(g\mu_B)^2}. \tag{A·8}$$

Since $\chi_k''(\omega_k)$ is expressed as[14]

$$\chi_k''(\omega_k) = (g\mu_B)^2\omega_k \,\mathrm{Re}\int_0^\infty (S^z_{-k}(t), S_k{}^z(0))\exp(-i\omega_k t)\,dt\,, \qquad (A\cdot 9)$$

we see that the expression of Eq. (A·8) is equal to the first term in Eq. (14).

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