Phonon-Magnon Interaction in Magnetic Crystals. II. Antiferromagnetic Systems*

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A theoretical study of phonon-magnon interaction in antiferromagnets is made on the basis of a microscopic mechanism developed earlier. The mechanism, which in essence takes into account the mixing of excited orbital states with the ground orbital states of the magnetic ions owing to crystal field oscillations, is applied to a crystal which can be subdivided into two interpenetrating identical sublattices coupled antiferromagnetically.

The interaction terms for one-phonon direct processes are first derived following the methods of the previous paper. The expressions for phonon-magnon relaxation times are then obtained for these processes in the low-temperature limit. It is found that the relaxation time τ_{sp} is inversely proportional to fifth power of temperature (T^5) in this region. Numerical estimate for MnF_2 at 10°K gives the tentative value of $\tau_{sp}\sim 10^{-8}$sec.

1. INTRODUCTION

In a previous paper,[1] a microscopic theory of phonon-magnon interactions in ferromagnetically coupled lattices was developed from first principles. The central theme of the theory consisted in taking into account the mixing of excited orbital states with the ground orbital states of the magnetic ions owing to crystal-field oscillations of appropriate symmetry. The relevant exchange Hamiltonian and interaction terms were obtained by making use of such one-electron perturbed states in the second quantization representation. This mechanism gave the right order of magnitude for phonon-magnon relaxation times in ferromagnetic systems.

The purpose of the present paper is to develop a similar theory of phonon-magnon interactions in antiferromagnetically coupled two-sublattice systems. Pincus and Winter[2] have phenomenologically discussed the effects of phonon-magnon interactions on nuclear spin-lattice relaxation rates of antiferromagnets. However, experiments aimed at correlating the linewidth of antiferromagnetic resonance (AFMR) absorption with the spin-lattice relaxation times are lacking. Some preliminary suggestions to explain the AFMR linewidth owing to fluctuations of the effective molecular field at the site of an individual spin have been made by Townes.[3]

Although the problem of the antiferromagnetic ground state of a three-dimensional network of spins has not been solved, the two-sublattice model, with spins in one pointing up and those of the other in the reverse direction can be regarded as representing the reality fairly closely.[4] Spin-wave theories for such two-sublattice antiferromagnets have been developed by several authors[4–9] by taking proper cognizance of the anisotropy energy. Recently, the study of magnon-magnon interactions in antiferromagnets has been carried out following the above two-sublattice spin-wave theory by Genkin and Fain.[10]

In what follows, we adopt a similar procedure in conjunction with the mechanism developed in SU(I) to study the phonon-magnon interaction in antiferromagnets. After formulating the interaction terms, the relaxation time for the establishment of equilibrium at low temperatures between the phonon and magnon systems for the one-phonon direct process is calculated. Two-phonon Raman processes are not considered in view of their negligible contribution at low temperatures as expected from the calculations of SU(I).

2. FORMULATION OF PHONON-MAGNON INTERACTION HAMILTONIAN

We consider two interpenetrating simple cubic sublattices of magnetic ions with one localized d electron. The spins on sublattice 1 point up, and those on sublattice 2 point down. The two together form a body-centered cubic structure of the magnetic system. Thus, the nearest neighbor of an ion belonging to sublattice 1 is on sublattice 2 with $z=8$ and vice versa. With the above model and following the procedure outlined in SU(I), we get the total Hamiltonian of the system including anisotropy terms as

$$H=H_{\mathrm{L}}+H_{\mathrm{el}}+H_{\mathrm{ex}}+H_{\mathrm{Z}}+H_{\mathrm{an}}+H_{\mathrm{int}}, \qquad (2.1)$$

where the symbols, respectively, stand for contributions to the total Hamiltonian due to lattice, one-electron terms, isotropic exchange, Zeeman, anisotropy, and

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1 K. P. Sinha and U. N. Upadhyaya, Phys. Rev. **127**, 432 (1962); hereafter referred to as SU(I).
2 P. Pincus and J. Winter, Phys. Rev. Letters **7**, 269 (1961); see also V. N. Kashcheev, Fiz. Tverd. Tela **4**, 755 (1962) [translation: Soviet Phys.—Solid State **4**, 556 (1962)].
3 F. M. Johnson and A. H. Nethercot, Jr., Phys. Rev. **114**, 705 (1959).
4 J. Van Kranendonk and J. H. Van Vleck, Rev. Mod. Phys. **30**, 1 (1958).
5 P. W. Anderson, Phys. Rev. **86**, 694 (1952).
6 R. Kubo, Phys. Rev. **87**, 568 (1952).
7 J. M. Ziman, Proc. Phys. Soc. (London) **65**, 540, 548 (1952).
8 T. Oguchi, Phys. Rev. **117**, 117 (1960).
9 J. Korringa, Phys. Rev. **125**, 1972 (1962).
10 V. N. Genkin and V. M. Fain, Zh. Eksperim. i Teor. Fiz. **41**, 1522 (1961) [translation: Soviet Phys.—JETP **14**, 1086 (1962)].

interaction terms. The explicit forms are given below:

$$H_{\mathrm{L}}=\sum_{\mathbf{q}p} \hbar\omega_{\mathbf{q}p}(b_{\mathbf{q}p}^{\dagger}b_{\mathbf{q}p}+\tfrac{1}{2}), \tag{2.2}$$

$$H_{\mathrm{el}}=\sum_{\alpha} E_{\alpha}'N_{\alpha}, \tag{2.3}$$

$$H_{\mathrm{ex}}=\tfrac{1}{2}\sum_{l,m} J(\mathbf{R}_{lm})P_{lm}^{\sigma}. \tag{2.4}$$

In Eq. (2.4) l runs over ions on sublattice 1 and m over sublattice 2 and $P_{lm}^{\sigma}=\frac{1}{2}+2\mathbf{S}_l\cdot\mathbf{S}_m$.

$$H_{\mathrm{Z}}=-Hg\mu_{\beta}\sum_{j=l,m} S_j^z. \tag{2.5}$$

H is the external magnetic field pointing in the z direction and the other symbols have their usual significance.

$$H_{\mathrm{an}}=-H_{\mathrm{A}}g\mu_{\beta}(\sum_l S_l^z-\sum_m S_m^z), \tag{2.6}$$

H_{A} being the anisotropy field.

$$H_{\mathrm{int}}=\sum_{l,m} 2\,{}^{\alpha}J(\mathbf{R}_{lm})\cdot\delta\mathbf{R}_h P_{lm}^{\sigma}+\text{higher order terms.} \tag{2.7}$$

In the above $\mathbf{S}_i$ represents the spin vector of the atom i, $\delta\mathbf{R}_h$ is the vector representing the change in the nearest-neighbor distance, $\omega_{\mathbf{q}p}$ is the mode branch frequency of lattice vibration, and $b_{\mathbf{q}p}^{\dagger}$, $b_{\mathbf{q}p}$ are the corresponding phonon creation and annihilation operators. Here $J(\mathbf{R}_{lm})$ represents the effective exchange integral. Although in SU(I) we expressed this in the Heisenberg formalism, it may be taken to include all other types of exchange or superexchange interaction terms.[11–14] As defined in SU(I),

$$\begin{aligned}{}^{\alpha}J(\mathbf{R}_{lm})=\sum_{\alpha}&\langle\phi_{\alpha}\phi_m|V_{12}|\phi_m\phi_l\rangle\\ &\times\langle\phi_{\alpha}|\mathbf{V}^h|\phi_l\rangle/(E_{\alpha}-E_l),\end{aligned} \tag{2.8}$$

with ϕ_{α} and $\phi_{l,m}$ standing for the excited- and ground-state orbitals, respectively, and $\mathbf{V}^h\equiv(\partial V/\partial\mathbf{R}_h)_0$, V being the crystal field due to the nearest-neighbor ions. Further, as before, we express $\delta\mathbf{R}_h$ in terms of $b_{\mathbf{q}p}^{\dagger}$, $b_{\mathbf{q}p}$ as

$$\delta\mathbf{R}_h=\frac{1}{\sqrt{N}}\sum_{\mathbf{q}p}\mathbf{g}_{\mathbf{q}p}\cdot(b_{\mathbf{q}p}^{\dagger}-b_{-\mathbf{q}p})(e^{i\mathbf{q}\cdot\mathbf{R}_{l0}}-e^{i\mathbf{q}\cdot\mathbf{R}_{m0}}), \tag{2.9}$$

with $\mathbf{g}_{\mathbf{q}p}=(-i)\mathbf{e}_{\mathbf{q}p}(\hbar/2\omega_{\mathbf{q}p}M)^{1/2}$, $\mathbf{e}_{\mathbf{q}p}$ being the polarization vector and M the mass of the ion. We now transform the spin-dependent parts of the Hamiltonian Eq. (2.1) in terms of the spin deviation operators of the two sublattices, which are expressed below:

$$\begin{aligned}S_l^{+}&=S_l^x+iS_l^y=(2S)^{1/2}(1-n_l/2S)^{1/2}a_l,\\ S_l^{-}&=S_l^x-iS_l^y=(2S)^{1/2}a_l^{\dagger}(1-n_l/2S)^{1/2},\\ S-S_l^z&=a_l^{\dagger}a_l=n_l(\text{the spin deviation}).\end{aligned} \tag{2.10}$$

[11] T. Kasuya, Progr. Theoret. Phys. (Kyoto) **16**, 45 (1956).
[12] P. W. Anderson, Phys. Rev. **115**, 2 (1959).
[13] S. Koide, K. P. Sinha, and Y. Tanabe, Progr. Theoret. Phys. (Kyoto) **22**, 647 (1959); K. P. Sinha, Indian J. Phys. **35**, 484 (1961).
[14] See P. O. Löwdin, Rev. Mod. Phys. **34**, 80 (1962), for other references.

Likewise, for the other sublattice

$$\begin{aligned}S_m^{+}&=(2S)^{1/2}d_m^{\dagger}(1-n_m/2S)^{1/2},\\ S_m^{-}&=(2S)^{1/2}(1-n_m/2S)^{1/2}d_m,\\ S+S_m^z&=n_m.\end{aligned} \tag{2.11}$$

These operators satisfy the commutation relations

$$aa^{\dagger}-a^{\dagger}a=1 \quad\text{and}\quad dd^{\dagger}-d^{\dagger}d=1.$$

Using Eqs. (2.10) and (2.11), the spin-dependent part of the Hamiltonian can be written as

$$\begin{aligned}H_s&=H_{\mathrm{ex}}+H_{\mathrm{Z}}+H_{\mathrm{an}}+H_{\mathrm{int}}\\ &=\text{constant}+\sum_{l,m} J(\mathbf{R}_{lm})S\{(a_ld_m+a_l^{\dagger}d_m^{\dagger}+a_l^{\dagger}a_l\\ &\quad+d_m^{\dagger}d_m)\}+g\mu_{\beta}\{H(\sum_l a_l^{\dagger}a_l-\sum_m d_m^{\dagger}d_m)\\ &\quad+H_{\mathrm{A}}(\sum_l a_l^{\dagger}a_l+\sum_m d_m^{\dagger}d_m)\}\\ &\quad+\frac{4S}{\sqrt{N}}\sum\sum_{l,m,\mathbf{q}p}\mathbf{g}_{\mathbf{q}p}\cdot{}^{\alpha}J(\mathbf{R}_{lm})[a_ld_m+a_l^{\dagger}d_m^{\dagger}+a_l^{\dagger}a_l\\ &\quad+d_m^{\dagger}d_m][b_{\mathbf{q}p}^{\dagger}-b_{-\mathbf{q}p}][e^{i\mathbf{q}\cdot\mathbf{R}_{l0}}-e^{i\mathbf{q}\cdot\mathbf{R}_{m0}}]\\ &\quad+\cdots.\end{aligned} \tag{2.12}$$

In writing (2.12) we have neglected the terms of the type $n_{l,m}/2S$ and higher order terms in the expansion of $(1-n_{l,m}/2S)^{1/2}$ and the constant includes the terms independent of spin-deviation operators.

The Hamiltonian H_s can be written in the spin-wave representation by making use of the Fourier transforms of the spin deviation operators given by

$$\begin{aligned}a_l&=(2/N)^{1/2}\sum_{\lambda}\exp(-i\boldsymbol{\kappa}_{\lambda}\cdot\mathbf{R}_1^0)a_{\lambda},\\ a_l^{\dagger}&=(2/N)^{1/2}\sum_{\lambda}\exp(i\boldsymbol{\kappa}_{\lambda}\cdot\mathbf{R}_1^0)a_{\lambda}^{\dagger},\\ d_m&=(2/N)^{1/2}\sum_{\lambda}\exp(i\boldsymbol{\kappa}_{\lambda}\cdot\mathbf{R}_m^0)d_{\lambda},\\ d_m^{\dagger}&=(2/N)^{1/2}\sum_{\lambda}\exp(-i\boldsymbol{\kappa}_{\lambda}\cdot\mathbf{R}_m^0)d_{\lambda}^{\dagger},\end{aligned} \tag{2.13}$$

where the propagation vector $\boldsymbol{\kappa}_{\lambda}$ runs over $N/2$ points of the first Brillouin zone of the reciprocal space of the lattice. With the help of (2.13) we can express (2.12) as

$$\begin{aligned}H_s&=\text{const}+\sum_{\lambda} zJ(\mathbf{R}_h^0)S\{\gamma_{\lambda}(a_{\lambda}d_{\lambda}+a_{\lambda}^{\dagger}d_{\lambda}^{\dagger})\\ &\quad+a_{\lambda}^{\dagger}a_{\lambda}+d_{\lambda}^{\dagger}d_{\lambda}\}+g\mu_{\beta}\{(H+H_A)\sum_{\lambda} a_{\lambda}^{\dagger}a_{\lambda}\\ &\quad+(H_A-H)\sum_{\lambda} d_{\lambda}^{\dagger}d_{\lambda}\}+\frac{4Sz}{\sqrt{N}}\sum_{\lambda}\sum_{\mathbf{q}p}\mathbf{g}_{\mathbf{q}p}\cdot{}^{\alpha}J(\mathbf{R}_h^0)\\ &\quad\times[(\gamma_{\lambda-\mathbf{q}}-\gamma_{\lambda})a_{\lambda}d_{\lambda-\mathbf{q}}+(\gamma_{\lambda+\mathbf{q}}-\gamma_{\lambda})a_{\lambda}^{\dagger}d_{\lambda+\mathbf{q}}^{\dagger}\\ &\quad+(1-\gamma_{\mathbf{q}})a_{\lambda}^{\dagger}a_{\lambda+\mathbf{q}}+(\gamma_{\mathbf{q}}-1)d_{\lambda}^{\dagger}d_{\lambda-\mathbf{q}}]\\ &\quad\times[b_{\mathbf{q}p}^{\dagger}-b_{-\mathbf{q}p}],\end{aligned} \tag{2.14}$$

where

$$\gamma_\lambda \equiv [\sum_h \exp(\pm i\boldsymbol{\kappa}_\lambda\cdot \mathbf{R}_h{}^0)]/z; \quad \mathbf{R}_h{}^0 = \mathbf{R}_l{}^0 - \mathbf{R}_m{}^0. \tag{2.15}$$

In the above, $J(\mathbf{R}_h{}^0)$ and ${}^\alpha J(\mathbf{R}_h{}^0)$ are assumed to be the same for all nearest-neighbor interactions; the summation over h extends to nearest neighbors. Further, in deriving (2.14) from (2.12) we have carried out the summation over l or m utilizing the following interference conditions, for the terms in the square brackets:

$$\boldsymbol{\kappa}_{\lambda'} - \boldsymbol{\kappa}_\lambda = \pm \mathbf{q}, \tag{2.16}$$

with the plus sign before $\mathbf{q}$ being used for the first and the last terms and the minus sign for the second and the third terms in the first square bracket of (2.14). It can be seen from (2.14) that the pure spin part of H_s, i.e., terms in the curly brackets, is not diagonal. To diagonalize the pure spin part as well as to write the interaction terms in the same representation, we make use of the following canonical transformation[15]:

$$\begin{aligned} a_\lambda &= \alpha_\lambda \cosh\theta_\lambda + \beta_\lambda{}^\dagger \sinh\theta_\lambda, \\ a_\lambda{}^\dagger &= \alpha_\lambda{}^\dagger \cosh\theta_\lambda + \beta_\lambda \sinh\theta_\lambda, \\ d_\lambda &= \alpha_\lambda{}^\dagger \sinh\theta_\lambda + \beta_\lambda \cosh\theta_\lambda, \\ d_\lambda{}^\dagger &= \alpha_\lambda \sinh\theta_\lambda + \beta_\lambda{}^\dagger \cosh\theta_\lambda, \end{aligned} \tag{2.17}$$

and

$$\tanh 2\theta_\lambda = -(\omega_e\gamma_\lambda/\omega_e + \omega_A), \tag{2.18}$$

and the symbols $\omega_e = 2zSJ(\mathbf{R}_h{}^0)/\hbar$ and $\omega_A = g\mu_\beta H_A/\hbar$. Using the magnon operators α, β, the pure magnon and interaction Hamiltonian in (2.14) take the forms given below.

$$H_{\rm m} = \sum_\lambda \hbar\omega_\lambda{}^+(\alpha_\lambda{}^\dagger\alpha_\lambda + \tfrac{1}{2}) + \sum_\lambda \hbar\omega_\lambda{}^-(\beta_\lambda{}^\dagger\beta_\lambda + \tfrac{1}{2}), \tag{2.19}$$

where

$$\omega_\lambda{}^\pm = [(\omega_A + \omega_e)^2 - \omega_e{}^2\gamma_\lambda{}^2]^{1/2} \pm \omega_H, \qquad \omega_H \equiv g\mu_\beta H/\hbar. \tag{2.19a}$$

The above for $\boldsymbol{\kappa}_\lambda = 0$ gives the well-known relation for AFMR frequency.[16] Likewise, the phonon-magnon interaction terms reduce to

$$\begin{aligned} H_{\rm int} = \sum_{\lambda\mathbf{q}p} [&A_{\lambda\mathbf{q}p}(\alpha_\lambda\alpha_{\lambda-\mathbf{q}}{}^\dagger b_{\mathbf{q}p}{}^\dagger - \alpha_\lambda{}^\dagger\alpha_{\lambda-\mathbf{q}} b_{\mathbf{q}p}) \\ &+ B_{\lambda\mathbf{q}p}(\alpha_\lambda\beta_{\lambda-\mathbf{q}} b_{\mathbf{q}p}{}^\dagger - \alpha_\lambda{}^\dagger\beta_{\lambda-\mathbf{q}}{}^\dagger b_{\mathbf{q}p}) \\ &+ A_{\lambda\mathbf{q}p}(\beta_\lambda\beta_{\lambda-\mathbf{q}}{}^\dagger b_{\mathbf{q}p} - \beta_\lambda{}^\dagger\beta_{\lambda-\mathbf{q}} b_{\mathbf{q}p}{}^\dagger)], \end{aligned} \tag{2.20}$$

where

$$\begin{aligned} A_{\lambda\mathbf{q}p} = \frac{4Sz\,{}^\alpha J(\mathbf{R}_h{}^0)\cdot \mathbf{g}_{\mathbf{q}p}}{\sqrt{N}} [&(\gamma_{\lambda-\mathbf{q}} - \gamma_\lambda)\sinh(\theta_{\lambda-\mathbf{q}} - \theta_\lambda) \\ &+ (1-\gamma_\mathbf{q})\cosh(\theta_{\lambda-\mathbf{q}} - \theta_\lambda)], \end{aligned} \tag{2.21}$$

$$\begin{aligned} B_{\lambda\mathbf{q}p} = \frac{4Sz\,{}^\alpha J(\mathbf{R}_h{}^0)\cdot \mathbf{g}_{\mathbf{q}p}}{\sqrt{N}} [&(\gamma_{\lambda-\mathbf{q}} - \gamma_\lambda)\cosh(\theta_{\lambda-\mathbf{q}} - \theta_\lambda) \\ &+ (1-\gamma_\mathbf{q})\sinh(\theta_{\lambda-\mathbf{q}} - \theta_\lambda)]. \end{aligned}$$

In deriving (2.20) from the interaction part of (2.14), we have omitted the terms which represent processes involving simultaneous creation or annihilation of two magnons and one phonon as they will not conserve energy.[17]

3. PHONON-MAGNON RELAXATION PROCESSES

Noting the properties of the boson creation and annihilation operators pertaining to transitions between different states in the occupation number representation [see SU(I)], the transition probabilities of the various processes contained in Eq. (2.20) can be easily written as

$$\begin{aligned} W(n_\lambda, n_{\lambda-\mathbf{q}}, N_{\mathbf{q}p} \to (n_\lambda - 1)(n_{\lambda-\mathbf{q}}+1)(N_{\mathbf{q}p}+1)) &= (2\pi/\hbar)|A_{\lambda\mathbf{q}p}|^2(n_\lambda)(n_{\lambda-\mathbf{q}}+1)(N_{\mathbf{q}p}+1) \\ &\quad\times\delta(E_{\lambda-\mathbf{q}} + E_\mathbf{q} - E_\lambda), \end{aligned} \tag{3.1a}$$

$$\begin{aligned} W(n_\lambda, n_{\lambda-\mathbf{q}}, N_{\mathbf{q}p} \to (n_\lambda + 1)(n_{\lambda-\mathbf{q}}-1)(N_{\mathbf{q}p}-1)) &= (2\pi/\hbar)|A_{\lambda\mathbf{q}p}|^2(n_\lambda+1)(n_{\lambda-\mathbf{q}})(N_{\mathbf{q}p}) \\ &\quad\times\delta(E_{\lambda-\mathbf{q}} + E_\mathbf{q} - E_\lambda), \end{aligned} \tag{3.1b}$$

$$\begin{aligned} W(n_\lambda, n_{\lambda-\mathbf{q}}', N_{\mathbf{q}p} \to (n_\lambda - 1)(n_{\lambda-\mathbf{q}}'-1)(N_{\mathbf{q}p}+1)) &= (2\pi/\hbar)|B_{\lambda\mathbf{q}p}|^2(n_\lambda)(n_{\lambda-\mathbf{q}}')(N_{\mathbf{q}p}+1) \\ &\quad\times\delta(E_\lambda + E_{\lambda-\mathbf{q}}' - E_\mathbf{q}), \end{aligned} \tag{3.2a}$$

$$\begin{aligned} W(n_\lambda, n_{\lambda-\mathbf{q}}', N_{\mathbf{q}p} \to (n_\lambda + 1)(n_{\lambda-\mathbf{q}}'+1)(N_{\mathbf{q}p}-1)) &= (2\pi/\hbar)|B_{\lambda\mathbf{q}p}|^2(n_\lambda+1)(n_{\lambda-\mathbf{q}}'+1)(N_{\mathbf{q}p}) \\ &\quad\times\delta(E_\lambda + E_{\lambda-\mathbf{q}}' - E_\mathbf{q}), \end{aligned} \tag{3.2b}$$

$$\begin{aligned} W(n_\lambda', n_{\lambda-\mathbf{q}}', N_{\mathbf{q}p} \to (n_\lambda' - 1)(n_{\lambda-\mathbf{q}}'+1)(N_{\mathbf{q}p}-1)) &= (2\pi/\hbar)|A_{\lambda\mathbf{q}p}|^2(n_\lambda')(n_{\lambda-\mathbf{q}}'+1)(N_{\mathbf{q}p}) \\ &\quad\times\delta(E_\lambda' + E_\mathbf{q} - E_{\lambda-\mathbf{q}}'), \end{aligned} \tag{3.3a}$$

$$\begin{aligned} W(n_\lambda', n_{\lambda-\mathbf{q}}', N_{\mathbf{q}p} \to (n_\lambda' + 1)(n_{\lambda-\mathbf{q}}'-1)(N_{\mathbf{q}p}+1)) &= (2\pi/\hbar)|A_{\lambda\mathbf{q}p}|^2(n_\lambda'+1)(n_{\lambda-\mathbf{q}}')(N_{\mathbf{q}p}+1) \\ &\quad\times\delta(E_\lambda' + E_\mathbf{q} - E_{\lambda-\mathbf{q}}'), \end{aligned} \tag{3.3b}$$

where n_λ, n_λ', $N_{\mathbf{q}p}$, respectively, represent the occupation numbers of magnon associated with energies $\hbar\omega_\lambda{}^+$, $\hbar\omega_\lambda{}^-$ and phonons of energy $\hbar\omega_{\mathbf{q}p}$. The δ functions ensure the conservation of energy. The rate of transfer of energy between the magnon and phonon systems is

[15] T. Nagamiya, K. Yoshida, and R. Kubo, in *Advances in Physics*, edited by N. F. Mott (Taylor and Francis, Ltd., London, 1955), Vol. 4, p. 1.

[16] C. Kittel, Phys. Rev. **82**, 565 (1951).

[17] It may be remarked that in the processes involving β and $\beta^\dagger$ [see Eq. (2.20)] the momentum conservation law for particles is not apparently satisfied. However, we are dealing with quasi-particles and the momentum of the particle should not be taken identically equal to $\hbar\boldsymbol{\kappa}_\lambda$. See J. M. Ziman, *Electrons and Phonons* (Clarendon Press, Oxford, 1960); G. H. Wannier, *Elements of Solid-State Theory* (Cambridge University Press, New York, 1959).

given by

$$\dot{Q}=\dot{Q}_{\alpha}+\dot{Q}_{\alpha\beta}+\dot{Q}_{\beta}$$
$$=\sum_{qp}[\langle\dot{N}_{qp}\rangle_{\alpha}+\langle\dot{N}_{qp}\rangle_{\alpha\beta}+\langle\dot{N}_{qp}\rangle_{\beta}]\hbar\omega_{qp}$$
$$=(2\pi/\hbar)\sum_{\lambda qp}\hbar\omega_{qp}[|A_{\lambda qp}|^2\{(n_\lambda)(n_{\lambda-q}+1)(N_{qp}+1)$$
$$-(n_\lambda+1)(n_{\lambda-q})(N_{qp})\}\delta(E_{\lambda-q}+E_q-E_\lambda)$$
$$+|B_{\lambda qp}|^2\{(n_\lambda)(n_{\lambda-q}')(N_{qp}+1)$$
$$-(n_\lambda+1)(n_{\lambda-q}'+1)(N_{qp})\}\delta(E_\lambda+E_{\lambda-q}'-E_q)$$
$$+|A_{\lambda qp}|^2\{(n_\lambda'+1)(n_{\lambda-q}')(N_{qp}+1)$$
$$-(n_\lambda')(n_{\lambda-q}'+1)(N_{qp})\}\delta(E_\lambda'+E_q-E_{\lambda-q}')]. \quad (3.4)$$

In proceeding further, we neglect the Zeeman energy contribution to the magnon energy, i.e., $\omega_\lambda^+=\omega_\lambda^-=\omega_\lambda$. Hence, (n_λ) and (n_λ') may be expressed by the same Bose distribution function $1/[\exp(E_\lambda/kT)-1]$.

As in SU(I), we define $\Delta T=T_s-T_1=T-T_1$ and making use of the Taylor expansion of terms containing $(T-\Delta T)$ in powers of ΔT and keeping only the first-order terms, we get

$$\dot{Q}=\frac{2\pi}{\hbar}\frac{\Delta T}{T^2}\sum_{\lambda qp}\frac{(\hbar\omega_{qp})^2}{k}F(\lambda\mathbf{q}p)$$
$$\times\{|A_{\lambda qp}|^2e^{E_\lambda/kT}\delta(E_{\lambda-q}+E_q-E_\lambda)$$
$$+|B_{\lambda qp}|^2e^{E_q/kT}\delta(E_{\lambda-q}+E_\lambda-E_q)$$
$$+|A_{\lambda qp}|^2e^{E_{\lambda-q}/kT}\delta(E_\lambda+E_q-E_{\lambda-q})\}, \quad (3.5)$$

where

$$F(\lambda\mathbf{q}p)\equiv\frac{1}{(e^{E_{\lambda-q}/kT}-1)(e^{E_\lambda/kT}-1)(e^{E_q/kT}-1)}. \quad (3.6)$$

We change the summation into integration, and use the Debye approximation for phonons, namely, $\omega_{qp}=k\theta_Dqa/\hbar$, a being the lattice constant and for magnons neglecting ω_A compared to ω_e

$$\hbar\omega_\lambda=\hbar\omega_e(1-\gamma_\lambda^2)^{1/2}\approx2JS\kappa_\lambda a(2z)^{1/2}\equiv k\theta_c\kappa_\lambda a,$$

where we have used the approximation $(\boldsymbol{\kappa}_\lambda\cdot\mathbf{R}_h^0)\ll1$. The above defines the parameter θ_c. Let us now consider the forms of the coefficients $|A_{\lambda qp}|^2$ and $|B_{\lambda qp}|^2$ under the approximation $(\boldsymbol{\kappa}_\lambda\cdot\mathbf{R}_h^0)\ll1$. We get [with $\mathbf{g}_{qp}=(\hbar/2\omega_{qp}M)^{1/2}$ as explained in SU(I)]:

$$|A_{\lambda qp}|^2=\frac{16S^2z^2}{N}(\hbar/2\omega_{qp}M)[^\alpha J(\mathbf{R}_h^0)]^2$$
$$\times[\tfrac{1}{2}\{(\gamma_{\lambda-q}-\gamma_\lambda)^2+(1-\gamma_q)^2\}\cosh2(\theta_{\lambda-q}-\theta_\lambda)$$
$$+\{(\gamma_{\lambda-q}-\gamma_\lambda)(1-\gamma_q)\}\sinh2(\theta_{\lambda-q}-\theta_\lambda)$$
$$+\tfrac{1}{2}\{(1-\gamma_q)^2-(\gamma_{\lambda-q}-\gamma_\lambda)^2\}], \quad (3.8)$$

where if we use

$$\tanh2\theta_\lambda=-(\omega_e\gamma_\lambda)/(\omega_e+\omega_A)\approx-\gamma_\lambda, \quad (3.9)$$
$$\cosh2(\theta_{\lambda-q}-\theta_\lambda)=(1-\gamma_{\lambda-q}\gamma_\lambda)/[(1-\gamma_{\lambda-q}^2)(1-\gamma_\lambda^2)]^{1/2}, \quad (3.10)$$
$$\sinh2(\theta_{\lambda-q}-\theta_\lambda)=(\gamma_\lambda-\gamma_{\lambda-q})/[(1-\gamma_{\lambda-q}^2)(1-\gamma_\lambda^2)]^{1/2}.$$

Substituting (3.10) into (3.8) and using the approximation $\gamma_\lambda\approx1-\kappa_\lambda^2a^2/z$ for $\boldsymbol{\kappa}_\lambda\cdot\mathbf{a}\ll1$, we get after making use of the relations $E_\lambda=k\theta_ca\kappa_\lambda$ and $E_q=k\theta_Daq$, etc.,

$$|A_{\lambda qp}|^2=\frac{16}{N}\left(\frac{\hbar}{2\omega_{qp}M}\right)S^2[^\alpha J(\mathbf{R}_h^0)]^2\frac{1}{k^4}$$
$$\times\Bigg\{\Bigg[\frac{1}{\theta_c^4}(E_\lambda^6-E_\lambda^4E_{\lambda-q}^2-E_\lambda^2E_{\lambda-q}^4+E_{\lambda-q}^6)$$
$$+\frac{1}{\theta_D^4}(E_q^4E_\lambda^2+E_q^4E_{\lambda-q}^2)-\frac{2}{\theta_D^2\theta_c^2}(E_q^2E_\lambda^4$$
$$+E_q^2E_{\lambda-q}^4-2E_q^2E_\lambda^2E_{\lambda-q}^2)\Bigg]\Big/4E_{\lambda-q}E_\lambda$$
$$+\frac{1}{2}\left[\frac{E_q^4}{\theta_D^4}-\frac{1}{\theta_c^4}(E_\lambda^4+E_{\lambda-q}^4-2E_\lambda^2E_{\lambda-q}^2)\right]\Bigg\}. \quad (3.11)$$

We get a similar expression for $|B_{\lambda qp}|^2$ except that the sign before the second square bracket in (3.11) is minus. It is interesting to recall that as in the case of ferromagnets [SU(I)], the dependence of $|A_{\lambda qp}|^2$ is of the fourth order in propagation vectors. However, the present expression [cf. Eq. (3.11)] is more involved than the corresponding expression for ferromagnets.

We discuss the integration of the three terms in (3.5), i.e., $\dot{Q}_\alpha$, $\dot{Q}_{\alpha\beta}$, and $\dot{Q}_\beta$ separately. Thus, we have

$$\dot{Q}_\alpha=\frac{2\pi}{\hbar}\frac{\Delta T}{T^2}\frac{1}{k}\left[\frac{Na^3}{8\pi^3}\right]^2\int|A_{\lambda qp}|^2E_q^2F(\lambda\mathbf{q}p)$$
$$\times e^{E_\lambda/kT}\delta(E_{\lambda-q}+E_q-E_\lambda)d\tau_\lambda d\tau_q, \quad (3.12)$$

where

$$d\tau_\lambda=\kappa_\lambda^2d\kappa_\lambda\sin\theta_\lambda d\theta_\lambda d\varphi_\lambda,\quad d\tau_q=q^2dq\sin\theta_qd\theta_qd\varphi_q.$$

Integrating over angle variables with the help of the δ function, which gives a factor proportional to $E_{\lambda-q}$, the above reduces to

$$\dot{Q}_\alpha=\frac{8\pi^3}{\hbar}\frac{\Delta T}{T^2}\frac{1}{k^3\theta_c^2a^2}\left[\frac{Na^3}{8\pi^3}\right]^2\int E_q^2|A_{\lambda qp}|^2$$
$$\times E_{\lambda-q}F(\lambda\mathbf{q}p)e^{E_\lambda/kT}\kappa_\lambda q\,d\kappa_\lambda dq. \quad (3.13)$$

This expression with the help of the dimensionless variables

$$\eta\equiv E_\lambda/kT\quad\text{and}\quad\xi\equiv E_q/kT$$

can be written as

$$\dot{Q}_\alpha = G\int_0^\infty \frac{\xi^2 d\xi}{(e^\xi-1)}\int_{\xi r}^\infty \frac{\Phi^+(\eta,\xi)e^\eta \eta d\eta}{(e^{\eta-\xi}-1)(e^\eta-1)}, \tag{3.14}$$

where

$$G=\frac{N}{\pi^3}\left(\frac{\hbar}{Mk}\right)\frac{\Delta T}{T^2}\frac{T^{10}}{\theta_D^2\theta_c^4}S^2[{}^\alpha J(\mathbf{R}_h^0)]^2, \tag{3.15}$$

$$\Phi^\pm(\eta\xi)=\Bigg\{\Bigg[\frac{1}{\theta_c^4}(8\eta^4\xi^2-16\eta^3\xi^3+14\eta^2\xi^4-6\eta\xi^5+\xi^6) + \frac{1}{\theta_D^4}(2\eta^2\xi^4-2\eta\xi^5+\xi^6) + \frac{2}{\theta_D^2\theta_c^2}(4\eta\xi^5-4\eta^2\xi^4-\xi^6)\Bigg]\Bigg/4\eta(\eta-\xi) \pm\frac{1}{2}\Bigg[\frac{\xi^4}{\theta_D^4}+\frac{1}{\theta_c^4}(4\eta\xi^3-4\eta^2\xi^2-\xi^4)\Bigg]\Bigg\}, \tag{3.16}$$

and $r=(\theta_c+\theta_D/2\theta_D$. For $\theta_c>\theta_D$, $r>1$; however, for $\theta_c<\theta_D$ we have to use $r=1$ to satisfy the δ function condition. For very low temperature limits the integral in (3.14) is easily evaluated for all the terms of (3.16). We have

$\dot{Q}_\alpha$(low-temperature limit)

$$=10^4 G\left[\frac{1.7}{\theta_c^4}+\frac{1.7}{\theta_D^4}-\frac{3.3}{\theta_D^2\theta_c^2}\right]. \tag{3.17}$$

Following the same procedure, we get, after making the appropriate use of the δ function while integrating, the values of $\dot{Q}_{\alpha\beta}$ and $\dot{Q}_\beta$. Summing all the three expressions, we can write $\dot{Q}$ as

$\dot{Q}$ (low-temperature limit)

$$=\frac{10^4 N}{\pi^3}\left(\frac{\hbar}{Mk}\right)\frac{\Delta T}{T^2}\frac{T^{10}}{\theta_D^2\theta_c^4}S^2[{}^\alpha J(\mathbf{R}_h^0)]^2 \times\left[\frac{4.7}{\theta_c^4}+\frac{5.3}{\theta_D^4}-\frac{9.6}{\theta_D^2\theta_c^2}\right]. \tag{3.18}$$

Relaxation Time for Equilibration

The relaxation time for phonon-magnon interaction τ_{sp} is expressed as [SU(I)]

$$\frac{1}{\tau_{sp}}=\frac{\dot{Q}(1/C_S+1/C_L)}{\Delta T}, \tag{3.19}$$

where C_L and C_S are, respectively, the lattice and the spin system specific heats. For the low-temperature limit we use

$$C_L=\frac{12\pi^4}{5}Nk\left(\frac{T}{\theta_D}\right)^3=234Nk\left(\frac{T}{\theta_D}\right)^3, \tag{3.20a}$$

and for a bcc antiferromagnet[4]

$$C_S=4Nk(T/\theta_c)^3. \tag{3.20b}$$

Substituting (3.20) and (3.18) into (3.19), we get

$$\frac{1}{\tau_{sp}}=\frac{10^4}{\pi^3}\left(\frac{\hbar}{Mk^2}\right)\frac{T^5}{\theta_D^2\theta_c^4}\left(\frac{\theta_c^3}{4}+\frac{\theta_D^3}{234}\right) \times\left(\frac{4.7}{\theta_c^4}+\frac{5.3}{\theta_D^4}-\frac{9.6}{\theta_D^2\theta_c^2}\right)S^2[{}^\alpha J(\mathbf{R}_h^0)]^2. \tag{3.21}$$

It is interesting to note that the above expression gives, in the low temperature limit, a simple law of the temperature dependence of the spin-lattice relaxation time in antiferromagnets, namely,

$$\tau_{sp}\propto 1/T^5. \tag{3.22}$$

4. ESTIMATES AND DISCUSSION

We shall now apply the foregoing theoretical analysis to some specific systems. Unfortunately, we cannot compare the theoretical estimates with any experimental value, in that none is available for any system.

The system which may closely approximate the model chosen, i.e., a body-centered cubic distribution of magnetic ions with each interpenetrating simple cubic lattice representing one of the two sublattices, is perhaps MnF_2 (body-centered-tetragonal structure) on which some AFMR experiments have been carried out. The estimated exchange field H_E for this system is of the order of 10^5 and $H_A\sim 10^3$ Oe.[10] Thus, our approximation of neglecting H_A in comparison with H_E is reasonable. If we estimate θ_c, following the Weiss approximation, its value for $T_N\sim 70°$K (the Néel temperature for MnF_2) turns out to be 30°K. A rough measure of θ_D can be obtained from the melting point of MnF_2, i.e., $T_M\approx 1129°$K by making use of the Lindemann relation[18]:

$$\theta_D=B(T_M/MV^{2/3})^{1/2}, \tag{4.1}$$

where M is the mean atomic weight and V is the mean atomic volume. Using a value of the constant[18] B equal to 115, we get $\theta_D\sim 250°$K. We get nearly the same value by using the formula[18]

$$\theta_D\approx(\hbar/k)(10a/9M\chi)^{1/2}, \tag{4.2}$$

where χ is the compressibility. The value of χ is determined from the data of Benedek and Kushida.[19] In estimating

$${}^\alpha J(\mathbf{R}_h^0)=\textstyle\sum_\alpha\langle\phi_\alpha|\mathbf{V}^h|\phi_l\rangle\langle\phi_\alpha\phi_m|V_{12}|\phi_m\phi_l\rangle/\Delta E_\alpha,$$

[18] M. Blackman, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1955), Vol. VII, Part I.

[19] G. B. Benedek and T. Kushida, Phys. Rev. **118**, 46 (1960).

the procedure is the same as discussed in SU(I). $\langle\phi_\alpha|\mathbf{V}^h|\phi_l\rangle$ is expected to be of the order of 1×10^{-3} dyn as shown from our previous study as well as the ligand field calculations on inorganic complexes.[20]

The exchange integral of the type $\langle\phi_\alpha\phi_m|V_{12}|\phi_m\phi_l\rangle$ may be estimated from the latest calculations of Freeman and Watson.[21] On the basis of their calculation a minimum value of the above integral can be taken to be of the order of 10^{-3} eV. Actually, for the present integral one may expect a larger value because of the extended nature of the excited orbital. Further, the superexchange effects are mainly responsible for the spin coupling in magnetic compounds such as MnF_2, and the excited orbitals are to be chosen on the magnetic ion centers or linear combination of atomic orbitals involving magnetic as well as nonmagnetic ions. Thus, using a value of $\Delta E_\alpha \equiv E_\alpha - E_0 \sim 10$ eV, we get ${}^\alpha J(\mathbf{R}_h{}^0)$ of the order of 1×10^{-7} dyn. If one identifies ${}^\alpha J(\mathbf{R}_h{}^0)$ with $(dJ/d\mathbf{R}_h{}^0)$, another estimate can be made from the data of Benedek and Kushida.[19] Using their values, namely,

$$(1/T_N)(\partial T_N/\partial P)=4.4\times10^{-6}/(\text{kg/cm}^2),$$
$$(1/a)(\partial a/\partial P)=-(0.45\times10^{-6})/(\text{kg/cm}^2),$$
$$(1/c)(\partial c/\partial P)=-(0.31\times10^{-6})/(\text{kg/cm}^2),$$

where P is the hydrostatic pressure, a and c being the lattice constants of MnF_2, we get ${}^\alpha J(\mathbf{R}_h{}^0)\sim dJ/d\mathbf{R}_h{}^0 \sim 6\times10^{-8}$ dyn. This is in rough agreement with the value noted above; however, to be on the safe side, we shall use a value of ${}^\alpha J(\mathbf{R}_h{}^0)$ to be of the order of 10^{-8} dyn. Thus, with $S=5/2$ and at $T=10°$K, we have

$$1/\tau_{\text{sp}} \approx 10^8 \text{ sec}^{-1};$$

i.e., τ_{sp} at 10°K is of the order of 10^{-8} sec. However, keeping in view the uncertainties in the values taken for the parameters involved, the above may be considered to be a tentative estimate.[22]

In contrast to the ferromagnetic case, an important difference is the absence of an exponential temperature factor in the expression for τ_{sp} in antiferromagnets which, in turn, is responsible for giving a shorter relaxation time in antiferromagnetic systems. This arises owing to the linear dispersion relation for antiferromagnets, namely, $\omega_\lambda \propto \kappa_\lambda$.

From the experimental results on the linewidth of AFMR on MnF_2 we expect the magnon-magnon relaxation time to be of the order of 10^{-10} sec at such low temperatures. Thus, the assumption, implicit in our theoretical analysis, that the magnons are in statistical equilibrium with each other would seem to be justified. It is desirable to have more experimental results on the AFMR of various antiferromagnets before a more quantitative estimate is attempted. Future experiments may help the verification of the temperature dependence suggested in the present work. We have already discussed the merits of choosing the present mechanism in preference to others in the previous paper [SU(I)], and the reasonable agreement between the experimental and calculated values of the relaxation time for ferromagnets impelled us to undertake a similar calculation for antiferromagnets.

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[20] A. D. Liehr and C. J. Ballhausen, Ann. Phys. (N. Y.) **3**, 304 (1958).

[21] A. J. Freeman and R. E. Watson, Phys. Rev. **124**, 1439 (1961).

[22] The expression for $(1/\tau_{\text{sp}})$ is very sensitive to the value of the parameter $\theta_c = 2JS(2z)^{1/2}/k$. If J is estimated from the exchange field (reference 10) $H_E \sim 6\times10^5$ Oe, θ_c would be of the order of 40°K. Using this value of θ_c, $\tau_{\text{sp}} \sim 10^{-7}$ sec at 10°K.