tion Λ_1 the single vector

$$\operatorname{Re}(\omega(\tau_x+i\tau_y);\,\omega^2(\tau_x+i\tau_y);\,(\tau_x+i\tau_y)),\quad (C1)$$

where the vectors at the three sites are written successively. This vector is determined uniquely. Equation (C1) describes a 1-dimensional configuration with three real vectors $\tau^{\rho\nu}$ for the directions at the different sites. Structures are therefore possible only for $\mathbf{k} = 0$, $\mathbf{k} = \mathbf{K}/2$ and $\mathbf{k} = \mathbf{K}/4$. The SSM is obeyed automatically because the representation is 1-dimensional and the conditions for the solutions at $\mathbf{K}/4$ are obeyed.

For Λ_2 the basis vectors can, e.g., be chosen as

$$(\tau_z; \tau_z; \tau_z)$$
 (C2a)

Im
$$(\omega(\tau_x + i\tau_y); \omega^2(\tau_x + i\tau_y); (\tau_x + i\tau_y));$$
 (C2b)

for k=0 one of them describes a ferromagnetic structure and the second a Yafet-Kittel²⁵ radial structure. The proper choice which diagonalizes the energy is obviously some mixture of these two configurations. All these mixtures obey the SSM because the representation is 1-dimensional. As the two vectors are real, the absolute phase can be chosen so that Eq. (36) is satisfied at all sites for $\mathbf{k} = \frac{1}{4}\mathbf{K}$.

For Λ_3 there are three pairs of solutions, e.g.,

$$(\omega^2 \tau_z; \omega \tau_z; \tau_z),$$
 (C3a)

$$((\tau_x+i\tau_y);(\tau_x+i\tau_y);(\tau_x+i\tau_y)),$$
 (C3b)

$$(\omega(\tau_x - i\tau_y); \omega^2(\tau_x - i\tau_y); (\tau_x - i\tau_y)), \quad (C3c)$$

²⁵ Y. Yafet and C. Kittel, Phys. Rev. 87, 290 (1952).

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for the first vector and, correspondingly,

$$(\omega \tau_z; \omega^2 \tau_z; \tau_z),$$
 (C4a)

$$(\boldsymbol{\tau}_x - i\boldsymbol{\tau}_y; \boldsymbol{\tau}_x - i\boldsymbol{\tau}_y; \boldsymbol{\tau}_x - i\boldsymbol{\tau}_y),$$
 (C4b)

$$(\omega^2(\tau_x+i\tau_y;\omega(\tau_x+i\tau_y);\tau_x+i\tau_y), \quad (C4c)$$

for the second vectors. We have chosen the vectors so as to diagonalize the rotations. The three vectors (C3) and (C4) transform under C_3 like $\tau_x + i\tau_y$ and $\tau_x - i\tau_y$, respectively. In general the configurations belonging to an irreducible representation will be combinations of these vectors and the right combinations depends on the interactions. The two vectors can, however, always be chosen so as to diagonalize C_3 and in this form they obey the SSM. On the other hand, Eqs. (36) and (38) will only be consistent with this choice in special cases. In general there are, therefore, no structures in the interior of the zone. When there are additional symmetry elements in the NMSG which do not add new sites, time inversion may cause two of the three repeated representations Λ_3 to be degenerate, and this gives enough freedom to allow one to construct spirals for arbitrary magnitudes of k.

We will not write down the vectors for six sites. The sites break up into two sets of three, each of which is permuted cyclically by the subgroup C_3 . For the 1-dimensional representations the SSM obviously holds. For Λ_3 one can again choose to diagonalize C_3 , and the vectors will then have the form of the τ^{ρ} of Eq. (C4) for each set separately, with different coefficients and magnitudes in each set. In general it is not possible to combine the two vectors belonging to one representation so as to satisfy the SSM at all sites.

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Phonon-Magnon Interaction in Magnetic Crystals*

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A microscopic theory of phonon-magnon interaction in magnetic crystals is developed from first principles. The crystal field oscillations are treated as perturbations which superpose some excited orbital states on the ground orbital state of the magnetic ions. When use is made of these perturbed states as the starting oneelectron functions in the second quantization representation, the formulation of the Heisenberg-type exchange interaction furnishes the relevant phonon-magnon interaction terms. Following the above interactions, the phonon-magnon relaxation times are calculated for the processes involving one-phonon direct and two-phonon Raman processes. Estimates made for iron, where the excited orbitals are taken to be the 4p and the ground $3d\gamma$ orbitals, yield values for the relaxation time for the one-phonon processes ($\tau_{sp} \approx 10^{-6}$ sec at 10°K) in agreement with the suggested results. Two-phonon Raman processes do not seem to be important at low temperatures.

INTRODUCTION

HE interaction between spin waves¹ and lattice vibrations is known to play an important role in the relaxation processes occurring in magnetic crystals,

particularly at low temperatures.² The first theoretical study was made by Akhiezer³ from a microscopic point of view by expanding the exchange and dipolar terms in power series with normal coordinates of the lattice

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^{*} Communication No. 491 from the National Chemical Laboratory, Poona-8, India. ¹ F. Bloch, Z. Physik 74, 295 (1932).

² J. Van Kranendonk and J. H. Van Vleck, Revs. Modern Phys. 30, 1 (1958). ³ A. Akhiezer, J. Phys. (U.S.S.R.) 10, 217 (1946).

vibrations. Polder,⁴ however, has pointed out some difficulties in his calculations. The recent studies of this problem are based on a phenomenological approach emanating from the macroscopic concepts of the exchange energy density and magnetoelastic coupling energy.⁵⁻⁸ Using this method, Kittel and Abraham⁵ and Kagnanov and Tsukernik⁷ have calculated the time for establishment of equilibrium between the phonon and magnon systems.

Inasmuch as the phenomenological approach does not provide a clear insight into the various microscopic processes involved, it is desirable to study the interaction on an atomic model from first principles.

In what follows, we consider the phonon-magnon interaction on an atomic model, taking into account the mixing of excited and ground state atomic orbitals due to crystal field oscillations. This procedure, when taken in conjunction with the two-electron operator. furnishes the relevant interaction Hamiltonian. We then calculate the relaxation times for the establishment of equilibrium between the spin and lattice systems arising through the direct and Raman processes involving phonons.

2. FORMULATION OF THE INTERACTION HAMILTONIAN

We choose a model consisting of magnetic atoms or ions, regularly arranged as in a crystal, each having in addition to a closed-shell ion core, one localized delectron. The effect of conduction electrons is unimportant as far as the phonon-magnon interaction is concerned.² The total Hamiltonian consists of the following parts:

$$H = H_e + H_L + H_C + H_Z, \qquad (2.1)$$

where H_e is the Hamiltonian involving one-electron coordinates, namely,

$$H_e = \sum_i p_i^2 / 2m + \sum_{i,n} U(\mathbf{r}_i - \mathbf{R}_n), \qquad (2.2)$$

 H_L is the lattice Hamiltonian expressed as

$$H_L = \sum_{qp} \hbar \omega_{qp} (b_{qp}^{\dagger} b_{qp} + 1/2), \qquad (2.3)$$

where b_{qp}^{\dagger} , b_{qp} standing for the phonon creation and annihilation operators pertaining to wave vector q and branch p,⁹ H_C is the electron-electron Coulomb

interaction operator, with the explicit form

$$H_{C} = \sum_{i \neq j} \frac{1}{2} \frac{e^{2}}{r_{ij}} \equiv \sum_{i \neq j} \frac{1}{2} g_{ij}, \qquad (2.4)$$

and H_Z is the Zeeman term.

In the above, $p_i^2/2m$ is the kinetic energy operator of the *i*th electron, $U(\mathbf{r}_i - \mathbf{R}_n)$ is its potential energy at \mathbf{r}_i due to the ion core at \mathbf{R}_n , ω_{qp} is the mode branch frequency of lattice vibration, and r_{ij} is the distance between electrons i and j. For the present, the spin-orbit interaction, anharmonic terms in lattice vibrations, and dipolar interactions between magnetic ions are not included. For the *i*th electron, the Hamiltonian (2.2) is rewritten as

$$H_{e}^{(i)} = p_{i}^{2}/2m + U(\mathbf{r}_{i} - \mathbf{R}_{l}^{0}) + V_{0} + H', \qquad (2.5)$$

$$H' = \sum_{h} \left(\frac{\partial V}{\partial \mathbf{R}_{h}}\right)_{0} \cdot \delta \mathbf{R}_{h} + \frac{1}{2} \sum_{h,h'} \left(\frac{\partial^{2} V}{\partial \mathbf{R}_{h} \partial \mathbf{R}_{h'}}\right) \delta \mathbf{R}_{h} \delta \mathbf{R}_{h'} + \cdots, \qquad (2.6)$$

$$V_{0} = \sum_{m \neq l} U(\mathbf{r}_{i} - \mathbf{R}_{m}^{0}); \quad \mathbf{R}_{h} = \mathbf{R}_{l} - \mathbf{R}_{m}.$$

Here $U(\mathbf{r}_i - \mathbf{R}_i)$ represents the potential acting on the electron when the ion cores are in undisplaced positions. V_0 is the static crystal field potential due to the nearestneighbor ions acting on the electron *i* when it is localized at the ion at \mathbf{R}_{l}^{0} . H' represents the first- and higher-order terms of the Taylor series development of the crystal field potential V of the neighboring ions in the relative displacements of the nearest-neighbor ions. We take the wavefunction $\phi_{a\sigma}$ as the solution of

$$[p^2/2m + U(\mathbf{r} - \mathbf{R}_l^0) + V_0]\phi_{a\sigma} = E_{a\sigma}\phi_{a\sigma}.$$
 (2.7)

The solution of (2.5) which includes H' as the perturbation and mixes some excited states with the ground-state functions is represented by

$$\psi_{a\sigma} = \phi_{a\sigma} + \sum_{\alpha} \frac{H_{\alpha a}' \phi_{\alpha \sigma}}{(E_{\alpha} - E_a)} + \cdots, \qquad (2.8)$$

where α represents the excited orbitals and the index σ stands for the spin state. With the above definition of one-electron functions, we can write the total Hamiltonian in occupation number representation by the method of second quantization as10

$$H = H_L + \sum_{a\sigma} E_{a\sigma'} A_{a\sigma}^{\dagger} A_{a\sigma} + \frac{1}{2} \sum_{a,b,c,d} \sum_{\sigma,\sigma'} A_{a\sigma}^{\dagger} A_{b\sigma'}^{\dagger} \times \langle a\sigma, b\sigma' | g_{12} | c\sigma, d\sigma' \rangle A_{c\sigma} A_{d\sigma'} + H_Z, \quad (2.9)$$

where $A_{a\sigma}^{\dagger}$, $A_{a\sigma}$ represent the creation and annihilation operators, respectively, for the one-electron states $\psi_{a\sigma}$

⁴ D. Polder, Phil. Mag. 40, 99 (1949).

⁵ C. Kittel and E. Abraham, Revs. Modern Phys. 25, 233 (1953).

 ⁶ C. Kittel, Phys. Rev. 110, 836 (1958).
 ⁶ M. I. Kaganov and V. M. Tsukernik, J. Exptl. Theoret. Phys. (U.S.S.R.) 36, 224 (1959) [translation Soviet Phys.—JETP 9, 151 (1959)].

⁸ For a more comprehensive bibliography see C. Kittel, Inter-national Conference on Magnetism and Crystallography, Kyoto, Japan (1961), Vol. 2, pp. 161–168. ⁹ J. M. Ziman, Electrons and Phonons (Clarendon Press, Oxford,

England, 1960).

¹⁰ L. D. Landau and E. M. Lifshitz, Quantum Mechanics (Pergamon Press, New York, 1958),

and obey the following relations:

$$A_{i}A_{k}^{\dagger} + A_{k}^{\dagger}A_{i} = \delta_{ik},$$

$$A_{i}A_{k} + A_{k}A_{i} = 0,$$

$$A_{i}^{\dagger}A_{i} = N_{i};$$

$$(2.10)$$

where k is the common index for both orbital and spin states, and N_i is the occupation number of the state *i*. For the ground occupied states $N_i=1$ and for empty excited states $N_i=0$. Further, for the orbital states which are occupied, the following relations hold between the above operators and the conventional spin operators:

$$A_{a}^{\dagger}(+)A_{a}(+)+A_{a}^{\dagger}(-)A_{a}(-) \equiv 1,$$

$$N_{a}(+)+N_{a}(-) \equiv 1,$$

$$N_{a}(+)-N_{a}(-) = 2S_{z}^{a}, \quad (2.11)$$

$$A_{a}^{\dagger}(+)A_{a}(-) = S_{+}^{a} = S_{x}^{a} + iS_{y}^{a},$$

$$A_{a}^{\dagger}(-)A_{a}(+) = S_{-}^{a} = S_{x}^{a} - iS_{y}^{a},$$

where (+) and (-) indicate two spin states of the electron.

Using relations (2.10) and (2.11) and summing over spin states, the Hamiltonian (2.9) can be written as

$$H = \sum_{qp} h \omega_{qp} (b_{qp}^{\dagger} b_{qp} + \frac{1}{2}) + \sum_{a} E_{a}' N_{a}$$

+ $\frac{1}{2} \sum_{l \neq m} J(R_{lm}) P_{lm}^{\sigma} + \frac{1}{2} \sum_{l \neq m,h} 4 \,^{\alpha} J(R_{lm}) P_{lm}^{\sigma} \delta \mathbf{R}_{h}$
+ $\frac{1}{2} \sum_{l \neq m} \sum_{hh'} \left[2 \,^{\alpha} J'(R_{lm}) + 2 \,^{\alpha\beta} J(R_{lm}) + 4 \,^{\alpha} J_{\beta}(R_{lm}) \right]$
 $\times P_{lm}^{\sigma} \delta \mathbf{R}_{h} \delta \mathbf{R}_{h'} + H_{Z}, \quad (2.12)$

where

$$P_{lm} = \frac{1}{2} + 2\mathbf{S}_{l} \cdot \mathbf{S}_{m},$$

$$J(R_{lm}) \equiv \langle \phi_{l}(1)\phi_{m}(2) | g_{12} | \phi_{m}(1)\phi_{l}(2) \rangle,$$

$$^{\alpha}J(R_{lm}) \equiv \sum_{\alpha} \langle \phi_{\alpha}\phi_{m} | g_{12} | \phi_{m}\phi_{l} \rangle \langle \phi_{\alpha} | V^{h} | \phi_{l} \rangle / (E_{\alpha} - E_{l}),$$

$$^{\alpha\beta}J(R_{lm}) \equiv \sum_{\alpha\beta} \frac{\langle \phi_{\alpha}\phi_{\beta} | g_{12} | \phi_{m}\phi_{l} \rangle \langle \phi_{\alpha} | V^{h} | \phi_{l} \rangle \langle \phi_{\beta} | V^{h'} | \phi_{m} \rangle}{(E_{\alpha} - E_{l})(E_{\beta} - E_{m})},$$

$$^{\alpha}J_{\beta}(R_{lm}) \equiv \sum_{\alpha\beta} \frac{\langle \phi_{\alpha}\phi_{m} | g_{12} | \phi_{\beta}\phi_{l} \rangle \langle \phi_{\alpha} | V^{h} | \phi_{l} \rangle \langle \phi_{\beta} | V^{h'} | \phi_{m} \rangle}{(E_{\alpha} - E_{l})(E_{\beta} - E_{m})},$$

$$^{\alpha}J'(R_{lm}) \equiv \sum_{\alpha} \langle \phi_{\alpha}\phi_{m} | g_{12} | \phi_{m}\phi_{l} \rangle \langle \phi_{\alpha} | V^{hh'} | \phi_{l} \rangle / (E_{\alpha} - E_{l}),$$

$$(2.13)$$

$$\mathbf{V}^{h} \equiv \left(\frac{\partial V}{\partial \mathbf{R}_{h}}\right)_{0}; \quad V^{hh'} \equiv \left(\frac{\partial^{2} V}{\partial \mathbf{R}_{h} \partial \mathbf{R}_{h'}}\right)_{0}; \quad \mathbf{R}_{lm} \equiv \mathbf{R}_{l}^{0} - \mathbf{R}_{m}^{0}.$$

The relative displacements $\delta \mathbf{R}_h$ between the ions at \mathbf{R}_l and R_m can be expressed in terms of the creation and annihilation operators of phonons⁹:

$$\delta \mathbf{R}_{h} = \frac{1}{\sqrt{N}} \sum_{\mathbf{q}p} (-i) \mathbf{e}_{\mathbf{q}p} (h/2\omega_{\mathbf{q}p}M)^{\frac{1}{2}} (b_{\mathbf{q}p}^{\dagger} - b_{-\mathbf{q}p}) \\ \times (e^{i\mathbf{q}\cdot\mathbf{R}_{l}0} - e^{i\mathbf{q}\cdot\mathbf{R}_{m}0}) \\ \equiv \frac{1}{\sqrt{N}} \sum_{\mathbf{q}p} \mathbf{g}_{\mathbf{q}p} (b_{\mathbf{q}p}^{\dagger} - b_{-\mathbf{q}p}) (e^{i\mathbf{q}\cdot\mathbf{R}_{l}0} - e^{i\mathbf{q}\cdot\mathbf{R}_{m}0}). \quad (2.14)$$

We now use the Holstein-Primakoff¹¹ (H.P.) spin deviation and other operators,

$$S_{+}{}^{l} = (2S)^{\frac{1}{2}} \left(1 - \frac{a_{l}{}^{\dagger}a_{l}}{2S}\right)^{\frac{1}{2}} a_{l},$$

$$S_{-}{}^{l} = (2S)^{\frac{1}{2}} a_{l}{}^{\dagger} \left(1 - \frac{a_{l}{}^{\dagger}a_{l}}{2S}\right)^{\frac{1}{2}},$$
 (2.15)

 $S - S_Z^l = a_l^{\dagger} a_l = n_l$ (the spin deviation),

where a_l^{\dagger} and a_l are the well-known creation and annihilation operators defined by H.P.¹¹ Following them, we neglect quantities of the type $\langle n_l \rangle/2S$ compared to unity. Further, simultaneously using the spin-wave Fourier transforms,

$$a_{\lambda} = (1/\sqrt{N}) \sum_{l} \exp(i\mathbf{k}_{\lambda} \cdot \mathbf{R}_{l}^{0}) a_{l},$$

$$a_{\lambda}^{\dagger} = (1/\sqrt{N}) \sum_{l} \exp(-i\mathbf{k}_{\lambda} \cdot \mathbf{R}_{l}^{0}) a_{l}^{\dagger},$$

$$a_{l} = (1/\sqrt{N}) \sum_{\lambda} \exp(-i\mathbf{k}_{\lambda} \cdot \mathbf{R}_{l}^{0}) a_{\lambda},$$

$$a_{l}^{\dagger} = (1/\sqrt{N}) \exp(i\mathbf{k} \cdot \mathbf{R}_{l}^{0}) a_{\lambda}^{\dagger},$$

(2.16)

where \mathbf{k}_{λ} is the reduced wave vector for quantized spin waves (magnons). We can express the phonon-magnon interaction terms contained in the Hamiltonian (2.12) as

$$H_{(\text{int})} = \frac{4}{\sqrt{N}} \sum_{h\lambda qp} 2S \,^{\alpha}J(\mathbf{R}_{h}) \left[e^{i\mathbf{k}_{\lambda} \cdot \mathbf{R}_{h}0} - e^{i(\mathbf{k}_{\lambda} - \mathbf{q}) \cdot \mathbf{R}_{h}0} + e^{-i\mathbf{q} \cdot \mathbf{R}_{h}0} - 1 \right] \mathbf{g}_{qp} a_{\lambda-q}^{\dagger} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{hh'} \sum_{qp,q'p'\lambda} \left[\alpha J'(\mathbf{R}_{h}^{0}) - 1 \right] \mathbf{g}_{qp} a_{\lambda-q}^{\dagger} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{hh'} \sum_{qp,q'p'\lambda} \left[\alpha J'(\mathbf{R}_{h}^{0}) - 1 \right] \mathbf{g}_{qp} a_{\lambda-q}^{\dagger} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{hh'} \sum_{qp,q'p'\lambda} \left[\alpha J'(\mathbf{R}_{h}^{0}) - 1 \right] \mathbf{g}_{qp} a_{\lambda-q}^{\dagger} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{hh'} \sum_{qp,q'p'\lambda} \left[\alpha J'(\mathbf{R}_{h}^{0}) - 1 \right] \mathbf{g}_{qp} a_{\lambda-q}^{\dagger} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{hh'} \sum_{qp,q'p'\lambda} \left[\alpha J'(\mathbf{R}_{h}^{0}) - 1 \right] \mathbf{g}_{qp} a_{\lambda-q}^{\dagger} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{hh'} \sum_{qp,q'p'\lambda} \left[\alpha J'(\mathbf{R}_{h}^{0}) - 1 \right] \mathbf{g}_{qp} a_{\lambda-q}^{\dagger} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{hh'} \sum_{qp,q'p'\lambda} \left[\alpha J'(\mathbf{R}_{h}^{0}) - 1 \right] \mathbf{g}_{qp} a_{\lambda-q}^{\dagger} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{hh'} \sum_{qp,q'p'\lambda} \left[\alpha J'(\mathbf{R}_{h}^{0} - b_{-qp}) \right] \mathbf{g}_{qp} a_{\lambda-q}^{\dagger} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{hh'} \sum_{qp,q'p'\lambda} \left[\alpha J'(\mathbf{R}_{h}^{0} - b_{-qp}) \right] \mathbf{g}_{qp} a_{\lambda-q}^{\dagger} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{hh'} \sum_{qp,q'p'\lambda} \left[\alpha J'(\mathbf{R}_{h}^{0} - b_{-qp}) \right] \mathbf{g}_{qp} a_{\lambda-q}^{\dagger} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{hh'} \sum_{qp,q'p'\lambda} \left[\alpha J'(\mathbf{R}_{h}^{0} - b_{-qp}) \right] \mathbf{g}_{qp} a_{\lambda-q}^{\dagger} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{hh'} \sum_{qp,q'p'\lambda} \left[\alpha J'(\mathbf{R}_{h}^{0} - b_{-qp}) \right] \mathbf{g}_{qp} a_{\lambda-q}^{\dagger} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{hh'} \sum_{qp,q'} \left[\alpha J'(\mathbf{R}_{h}^{0} - b_{-qp}) \right] \mathbf{g}_{qp} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{qp} \sum_{qp} \left[\alpha J'(\mathbf{R}_{h}^{0} - b_{-qp}) \right] \mathbf{g}_{qp} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{qp} \sum_{qp} \left[\alpha J'(\mathbf{R}_{h}^{0} - b_{-qp}) \right] \mathbf{g}_{qp} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{qp} \sum_{qp} \left[\alpha J'(\mathbf{R}_{h}^{0} - b_{-qp}) \right] \mathbf{g}_{qp} a_{\lambda} (b_{qp}^{\dagger} - b_{-qp}) + \frac{4}{N} \sum_{qp} \sum_{qp} \left[\alpha J'(\mathbf{R}_{h}^{0} - b_{-qp}) \right] \mathbf{g}_{qp} a_{\lambda} (b_{qp$$

$$+\alpha\beta J(\mathbf{R}_{h}^{0})+2\alpha J_{\beta}(\mathbf{R}_{h}^{0})][e^{i\mathbf{k}_{\lambda}\cdot\mathbf{R}_{h}0}-e^{i(\mathbf{k}_{\lambda}-\mathbf{q})\cdot\mathbf{R}_{h}0}+e^{-i\mathbf{q}\cdot\mathbf{R}_{h}0}+e^{-i\mathbf{q}\cdot\mathbf{R}_{h}\cdot\mathbf{0}}-1+e^{i\{(\mathbf{k}_{\lambda}-\mathbf{q})\cdot\mathbf{R}_{h}0-\mathbf{q}\cdot\mathbf{R}_{h}\cdot\mathbf{0}\}}-e^{i(\mathbf{k}_{\lambda}\cdot\mathbf{R}_{h}0-\mathbf{q}\cdot\mathbf{R}_{h}\cdot\mathbf{0})}$$

$$-e^{-i(\mathbf{q}\cdot\mathbf{R}_{h}0+\mathbf{q}\cdot\mathbf{R}_{h}'0)}]\mathbf{g}_{\mathbf{q}\cdot\mathbf{p}}\mathbf{g}_{\mathbf{q}\cdot\mathbf{p}'}a_{\lambda-\mathbf{q}-\mathbf{q}'}^{\dagger}a_{\lambda}(b_{\mathbf{q}\cdot\mathbf{p}}^{\dagger}-b_{-\mathbf{q}\cdot\mathbf{p}})(b_{\mathbf{q}'\cdot\mathbf{p}'}^{\dagger}-b_{-\mathbf{q}'\cdot\mathbf{p}'})+\cdots.$$
(2.17)

In arriving at (2.17), the summation over l has been carried out utilizing the relation⁹

$$\sum_{l} e^{i\mathbf{S} \cdot \mathbf{R} l^{0}} = 0 \quad \text{for} \quad \mathbf{S} \neq \mathbf{K},$$

= N for $\mathbf{S} = \mathbf{K},$ (2.18)

where K is the vector of the reciprocal lattice including zero and S stands for the phonon or magnon wave vectors. It may be noted that only normal processes,

¹¹ T. Holstein and H. Primakoff, Phys. Rev. 58, 1098 (1940).

here

i.e., $\mathbf{K}=0$, are under consideration here. The various exchange integrals occurring in (2.17) are taken for the nearest neighbors and hence, $\mathbf{R}_{h}^{0} = (\mathbf{R}_{l}^{0} - \mathbf{R}_{m}^{0})$, where l and m are suffixes for the nearest neighbors. Finally, the following momentum conservation relations between the magnon and phonon wave vectors may be noted which hold, respectively, for the one- and twophonon processes and have already been utilized in deriving (2.17):

$$\mathbf{k}_{\lambda}' + \mathbf{q} - \mathbf{k}_{\lambda} = 0, \qquad (2.19)$$

$$\mathbf{k}_{\lambda}' + \mathbf{q} + \mathbf{q}' - \mathbf{k}_{\lambda} = 0. \tag{2.20}$$

The first term in (2.17) represents the one-phonon direct process which involves the creation or annihilation of a phonon of wave vector \mathbf{q} accompanied with the scattering of two magnons (with wave vectors \mathbf{k}_{λ} and \mathbf{k}_{λ}'). The second term contains Raman processes involving the creation and annihilation of two phonons (wave vectors \mathbf{q} and \mathbf{q}'). In the following sections we consider the effect of one-phonon and two-phonon processes in the establishment of equilibrium between magnons and phonons.

3. EQUILIBRATION BETWEEN MAGNONS AND PHONONS

(i) One-Phonon Process

We calculate the transition probabilities of the various processes connecting the initial and final states using the well-known time-dependent perturbation expression,

$$W_{if} = (2\pi/\hbar) |H_p|_{if}^2 \delta(E_i - E_f).$$
 (3.1)

It is expedient to note the properties of the creation and annihilation operators pertaining to the magnon and phonon systems:

$$a_{\lambda}^{\dagger} | n_{\lambda} \rangle = (n_{\lambda} + 1)^{\frac{1}{2}} | n_{\lambda} + 1 \rangle,$$

$$a_{\lambda} | n_{\lambda} \rangle = n_{\lambda}^{\frac{1}{2}} | n_{\lambda} - 1 \rangle,$$

$$b_{qp}^{\dagger} | N_{qp} \rangle = (N_{qp} + 1)^{\frac{1}{2}} | N_{qp} + 1 \rangle,$$

$$b_{qp} | N_{qp} \rangle = N_{qp}^{\frac{1}{2}} | N_{qp} - 1 \rangle.$$
(3.2)

 $|\cdots n_{\lambda} \cdots \rangle$ represents the eigenket of the unperturbed magnon Hamiltonian,

$$= c + \sum_{\lambda} A_{\lambda} a_{\lambda}^{\dagger} a_{\lambda}, \qquad (3.2)$$

$$A_{\lambda} = \sum_{h} 2J(R_{h})S(e^{i\mathbf{k}_{\lambda}\cdot\mathbf{R}_{h}^{0}}-1) + 2\mu_{\beta}H,$$

 H_m

and μ_{β} and **H** are the Bohr magneton and the external magnetic field; the last term is the Zeeman term.

The magnon-dependent eigenvalues for $\mathbf{k}_{\lambda} \cdot \mathbf{R}_{h^0} \ll 1$ and for cubic crystals are

$$E_m = \sum_{\lambda} (2JSk_{\lambda}^2 a^2 + 2\mu_{\beta}H)(n_{\lambda} + 1/2), \qquad (3.3)$$

which gives us the dispersion relation, apart from a constant term involving H:

$$\hbar\omega_{\lambda} = k\theta_c a^2 k_{\lambda}^2, \qquad (3.4)$$

with $\theta_c \equiv 2JS/k$, k the Boltzmann constant, and a the nearest-neighbor distance. Likewise, $|\cdots N_{qp}\cdots\rangle$ represents the eigenkets of the lattice Hamiltonian (2.3). For eigenkets of the combined ground-state Hamiltonian (2.3) and (3.2), we use the notation

$$|\cdots n_{\lambda} \cdots; \cdots N_{qp} \cdots \rangle.$$
 (3.5)

Using (3.1), (3.2), and (3.5), the transition probability for the emission and absorption of a phonon can be expressed as

$$W(n_{\lambda-q}, n_{\lambda}, N_{qp} \rightarrow (n_{\lambda-q}+1), (n_{\lambda}-1), (N_{qp}+1)) = (2\pi/\hbar) |\Phi_{\lambda qp}|^2 (n_{\lambda-q}+1) (n_{\lambda}) (N_{qp}+1) \delta(E_{\lambda-q}+E_{qp}-E_{\lambda}), \quad (3.6)$$

 $W(n_{\lambda-\mathfrak{q}}, n_{\lambda}, N_{-\mathfrak{q}p} \to (n_{\lambda-\mathfrak{q}}+1), (n_{\lambda}-1), (N_{-\mathfrak{q}p}-1)) = (2\pi/\hbar) |\Phi_{\lambda\mathfrak{q}p}|^2 (n_{\lambda-\mathfrak{q}}+1) (n_{\lambda}) (N_{-\mathfrak{q}p}) \delta(E_{\lambda-\mathfrak{q}}-E_{-\mathfrak{q}}-E_{\lambda}), \quad (3.7)$

where

$$\Phi_{\lambda \mathfrak{q} p} = (4/\sqrt{N}) \sum_{h} 2S \, ^{\alpha} J(R_{h}) \left[e^{i\mathbf{k}_{\lambda} \cdot \mathbf{R}_{h}^{0}} - e^{i(\mathbf{k}_{\lambda} - \mathbf{q}) \cdot \mathbf{R}_{h}^{0}} - 1 + e^{-i\mathbf{q} \cdot \mathbf{R}_{h}^{0}} \right] \mathbf{g}_{\mathfrak{q} p}.$$

$$(3.8)$$

The transfer of energy between the magnon and phonon systems is expressed as

$$\dot{Q} = \sum_{qp} \hbar \omega_{qp} \langle \dot{N}_{qp} \rangle$$

$$= \frac{2\pi}{\hbar} \sum_{qp\lambda} |\Phi_{\lambda qp}|^2 \hbar \omega_{qp} [(n_{\lambda-q}+1)(n_{\lambda})(N_{qp}+1) - (n_{\lambda-q})(n_{\lambda}+1)(N_{qp})] \delta(E_{\lambda-q}+E_{qp}-E_{\lambda}). \tag{3.9}$$

The δ functions in (3.6) to (3.9) ensure the energy conservation. Let T_s and T_l be the spin and lattice temperatures, respectively, which govern the equilibrium Bose distribution of magnons and phonons. Thus with $T_s = T$ and $\Delta T = T_s - T_l$, we can write (3.9) after developing the terms containing $(T - \Delta T)$ in terms of Taylor series in powers of ΔT . Keeping only first-order terms, we get

$$\dot{Q} = \frac{2\pi}{\hbar} \frac{\Delta T}{T^2} \sum_{q,p\lambda} |\Phi_{\lambda q,p}|^2 \frac{(\hbar \omega_{q,p})^2}{k} \frac{e^{E_{\lambda}/kT}}{(e^{E_{\lambda-q}/kT} - 1)(e^{E_{\lambda}/kT} - 1)(e^{E_{q,p}/kT} - 1)} \delta(E_{\lambda-q} + E_{q,p} - E_{\lambda}).$$
(3.10)

We change the summation into integration, and use the Debye approximation for the dispersion relation of phonons, i.e., $\omega_{qp} = v_s |\mathbf{q}|$, where v_s is the velocity of sound. Further, $\Phi_{\lambda qp}$ can be simplified using the approximations¹²:

$$\begin{pmatrix} \frac{\partial V}{\partial \mathbf{R}_{h}} \cdot \delta \mathbf{R}_{h} \end{pmatrix} \approx \left| \frac{\partial V}{\partial \mathbf{R}_{h}} \right| |\delta \mathbf{R}_{h}|, \quad |e_{qp}| \sim 1,$$

$$\mathbf{g}_{qp} \approx (\hbar/2\omega_{qp}M)^{\frac{1}{2}},$$

$$\Phi_{\lambda qp} \approx \frac{4}{\sqrt{N}} \left(\frac{\hbar}{2\omega_{qp}M} \right)^{\frac{1}{2}} (i) (-2k_{\lambda}qa^{2}\cos\theta_{\lambda q}) [2S \ ^{\alpha}J(R_{h})].$$
(3.11)

and hence

Thus, (3.10) becomes

$$\dot{Q} = \frac{2\pi}{\hbar} \frac{2^8}{N} \frac{\Delta T}{T^2} S^2 [\alpha J(R_h)]^2 \left(\frac{\hbar^3 v_S}{2Mk}\right) a^4 \left(\frac{Na^3}{(2\pi)^3}\right)^2 \int q(k_\lambda^2 q^2 \cos^2\theta_{\lambda q}) \frac{e^{E_{\lambda/kT}}}{(e^{E_{\lambda-q/kT}}-1)(e^{E_{\lambda/kT}}-1)(e^{E_{\lambda/kT}}-1)} \times \delta(E_{\lambda-q}+E_{qp}-E_{\lambda}) d\tau_{\lambda} d\tau_q, \quad (3.12)$$

where

$$d\tau_{\lambda} = k_{\lambda}^{2} dk_{\lambda} \sin\theta_{\lambda} d\theta_{\lambda} d\varphi_{\lambda},$$

$$d\tau_{q} = q^{2} dq \sin\theta_{q} d\theta_{q} d\varphi_{q}.$$

In proceeding further we make use of the δ -function relation, $E_{\lambda-q} + E_{qp} - E_{\lambda} = 0,$

i.e.,

$$2k_{\lambda}q\theta_{c}ka^{2}\left(\frac{q}{2k_{\lambda}}-\cos\theta_{\lambda q}+\frac{\theta_{D}}{2\theta_{c}k_{\lambda}a}\right)=0.$$
 (3.13)

with

where

$$\theta_D = \hbar v_s / ka.$$

Making use of the conditions

$$\cos^{2}\theta_{\lambda} + \cos^{2}\theta_{q} + f^{2} - 2f \cos\theta_{\lambda} \cos\theta_{q} \leq 1,$$
$$f = \frac{1}{2k_{\lambda}} \left(q + \frac{\theta_{D}}{\theta_{c}a} \right),$$

which follows simply from (3.13), we integrate over the angular variables utilizing the δ -function property³.

The final result is written in terms of the variables

$$\eta = E_{\lambda}/kT = \theta_c a^2 k_{\lambda}^2/T$$

and

$$\xi = E_{qp}/kT = a\theta_D q/T$$

Thus

$$\dot{Q} = D \int_{0}^{\infty} d\xi \int_{(\xi+\beta)^{2}/4\beta}^{\infty} d\eta \ \xi^{4} \frac{(\xi+\beta)^{2}}{4} \times \frac{e^{\eta}}{(e^{\eta-\xi}-1)(e^{\eta}-1)(e^{\xi}-1)}, \quad (3.14)$$
where

$$D = \frac{4N}{\pi^3} \left(\frac{\hbar}{Mk}\right) \frac{\Delta T}{T^2} \frac{T^8}{\theta_D{}^6 \theta_c{}^2} S^2 [\alpha J(R_h)]^2,$$

$$\beta \equiv \theta_D{}^2/\theta_c T. \qquad (3.15)$$

¹² R. D. Mattuck and M. W. P. Strandburg, Phys. Rev. 119, 1204 (1960).

For integrating (3.14), we follow closely the method suggested earlier,³ and find the values for the limiting cases of low and high temperatures.

Low-Temperature Limit $(\beta \gg 1)$

$$Q = \frac{2^9 \times 3N}{\pi^3} \left(\frac{\hbar}{Mk}\right) \frac{\Delta T}{T^2} \frac{T^6}{\theta_D{}^2\theta_c{}^4} \\ \times \exp(-\beta/4) S^2 [\alpha J(R_h)]^2. \quad (3.16)$$

The exponential decrease of energy transfer with decreasing temperature in (3.16) is the same as that obtained by Akhierzer³ and others⁷; however, there are important differences in other factors.

High-Temperature Limit $(\beta \rightarrow 0)$

In integrating (3.14) for this case, the upper limit for ξ is taken as θ_D/T and in the final expression the terms in (θ_D/T) higher than two are neglected. We get

$$\dot{Q} = \frac{2N}{3\pi^3} \left(\frac{\hbar}{Mk}\right) \Delta T \left(\frac{\theta_D}{\theta_c^2}\right)^2 S^2 [\alpha J(R_h)]^2.$$
(3.17)

It may be noted that \dot{Q} in the higher temperature limit becomes independent or temperature in our mechanism.

Two-Phonon Process

The second term in (2.17) constitutes the two-phonon interaction Hamiltonian. In this, however, the terms representing the creation or annihilation of two phonons are relatively unimportant.¹³ Accordingly, these are neglected. We consider the other two terms which represent the Raman processes involving phonons. Thus the energy transfer owing to the two-phonon

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¹³ I. Waller, Z. Physik 79, 370 (1932).

Raman process can be taken as

$$\begin{split} \dot{Q}_{R} &= \sum_{\mathbf{q}p} \hbar \omega_{\mathbf{q}p} \langle \dot{N}_{\mathbf{q}p} \rangle \\ &= \frac{2\pi}{\hbar} \sum_{\mathbf{q}p,\mathbf{q}'p'} \hbar \omega_{\mathbf{q}p} |\sum_{hh'} \Phi_{\lambda qq'}{}^{hh'}|^{2} [(n_{\lambda-q-q'}+1)(n_{\lambda})(N_{\mathbf{q}p}+1)(N_{-q'p'}) \\ &- (n_{\lambda-q-q'})(n_{\lambda}+1)(N_{\mathbf{q}p})(N_{-q'p'}+1)] \delta(E_{\lambda-q-q'}+E_{q}-E_{\lambda}-E_{-q'}). \end{split}$$
(3.18)

As before, we define $\Delta T = T_s - T_l = T - T_l$ and, making use of the Taylor series expansion, we get for h = h'

$$\dot{Q}_{R} = \frac{2\pi}{\hbar} \sum_{\lambda qq'} \left(\frac{\hbar^{2}}{k} \right) \frac{\Delta T}{T^{2}} \sum_{h} \Phi_{\lambda qq'}^{hh} \frac{\omega_{qp}}{\omega_{qp}} (\omega_{qp} - \omega_{-q'p'}) \frac{e^{(E_{\lambda} + E_{-q'p'})/kT}}{(e^{E_{\lambda-q-q'}/kT} - 1)(e^{E_{\lambda/kT}} - 1)(e^{E_{qp}/kT} - 1)(e^{E_{-q'p'}/kT} - 1)} \times \delta(E_{\lambda-q-q'} + E_{qp} - E_{\lambda} - E_{-q'p'}). \quad (3.19)$$

The explicit form for $\sum_{h} \Phi_{\lambda qq'}{}^{hh}$ can be obtained by expanding the second term of (2.17) up to fourth order, i.e.,

$$\sum_{h} \Phi_{\lambda qq'}{}^{hh} = \frac{4}{N} \Big[{}^{\alpha}J'(R_{h}^{0}) + {}^{\alpha\beta}J(R_{h}^{0}) + 2{}^{\alpha}J_{\beta}(R_{h}^{0}) \Big] \times \frac{\hbar}{M} \frac{a^{4}}{(\omega_{qx}\omega_{-q'p'})^{\frac{1}{2}}} \frac{1}{6} \Big[k_{\lambda}{}^{2}qq'\cos\theta_{qq'} - k_{\lambda}q^{2}q'\cos\theta_{\lambda q'} - k_{\lambda}qq'^{2}\cos\theta_{\lambda q}} + 2k_{\lambda}{}^{2}qq'\cos\theta_{\lambda q}\cos\theta_{\lambda q'} - 2k_{\lambda}q^{2}q'\cos\theta_{\lambda q}q'\cos\theta_{\lambda q'} \cos\theta_{\eta q'} \Big].$$
(3.20)

One can change from summation to integration; however, a rigorous integration is hopelessly complicated owing to the occurrence of several angle variables in the δ function as well as $|\sum_{h} \Phi_{hqq'}{}^{hh}|^2$. One can at best attempt to give an order-of-magnitude estimate for certain specific conditions. We estimate (3.19) for the condition when a high-energy magnon is almost completely annihilated, creating a high-energy phonon and a very low energy magnon as the result of a collision with a very low energy phonon. The only significant term in the square bracket in (3.20) then is of the type $k_\lambda qq'^2 \cos\theta_{\lambda q} \sim k_\lambda qq'^2$.

We get, apart from numerical factors,

where

$$\dot{Q}_{R} \approx \frac{1}{\pi^{5}} \frac{N\hbar^{3}}{M^{2}k^{2}} \frac{\Delta T}{T^{2}} \left(\frac{T}{\theta_{D}}\right)^{12} \left(\frac{T}{\theta_{c}}\right)^{\frac{5}{2}} \frac{1}{\theta_{D}} S^{2} J_{R}^{2},$$
 (3.21)

$$J_R \equiv \left[{}^{\alpha}J'(R_h^0) + {}^{\alpha\beta}J(R_h^0) + 2{}^{\alpha}J_{\beta}(R_h^0) \right].$$
(3.22)

Likewise, when we consider an interaction which involves a small transfer of energy between the phonon and magnon system, i.e., for the term $k_{\lambda}^2 q q' \cos \theta_{qq'} \sim k_{\lambda}^2 q q'$, we get

$$\dot{Q}_{R} \approx \frac{1}{\pi^{5}} \frac{N\hbar^{3}}{M^{2}k^{2}} \frac{\Delta T}{T^{2}} \left(\frac{T}{\theta_{D}}\right)^{10} \left(\frac{T}{\theta_{c}}\right)^{7/2} \frac{1}{\theta_{D}} S^{2} J_{R}^{2}.$$
 (3.23)

4. RELAXATION TIME FOR EQUILIBRATION

We now proceed to calculate the relaxation time for the establishment of equilibrium between the magnon and phonon systems owing to the one-phonon and twophonon processes discussed in the preceding sections. The relevant expression is given by⁵

$$\frac{1}{\tau_{sp}} = \frac{Q(1/C_s + 1/C_l)}{\Delta T},$$
(4.1)

where τ_{sp} is the relaxation time; C_s and C_l are the specific heats of the spin and lattice systems, respectively. The following explicit forms for these are taken^{3,14} for the low-temperature limit:

$$C_{s} = \frac{15}{32} \frac{1}{\pi^{\frac{3}{2}}} k N \left(\frac{T^{3/2}}{\theta_{c}} \right) ; \quad C_{l} = \frac{12\pi^{4}}{5} k N \left(\frac{T}{\theta_{D}} \right)^{3} . \quad (4.2)$$

Using the expressions for the \dot{Q} 's in the preceeding section, the various relaxation times are given by:

One-Phonon Process

(i) Low-Temperature Limit $(\beta \gg 1)$

$$\frac{1}{\tau_{sp}} = \frac{3 \times 2^9}{\pi^3} \left(\frac{\hbar}{Mk^2}\right) \frac{T^4}{\theta_D^{2\theta} c^4} \left(\frac{\theta_c}{T}\right)^{\frac{3}{2}} \left(11 + \frac{\beta^{\frac{3}{4}}}{234}\right) \\ \times \exp[-\beta/4) S^2 [\alpha J(R_h^0)]^2, \quad (4.3)$$

(ii) High-Temperature Limit $(\beta \rightarrow 0)$

In the high-temperature limit, the specific heat of the lattice reduces to the well-known expression 3Nk. For the spin system, however, the analogous expression has not been suggested so far. The expression for C_s given in (4.2) is valid only at low temperature. As in the case of crystal lattice vibrations, if we take the upper limit of the magnon wave vectors as 1/a, then for the high-temperature limit one arrives at the expression for the specific heat of the spin system as $C_s = Nk/6\pi^2$, which is much smaller than the lattice specific heat at high temperatures. It is interesting to note that the characteristic temperature for the spin system implicit in the above assumption is the same as the temperature

¹⁴ C. Kittel, Solid-State Physics (John Wiley & Sons, Inc., New York, 1956), 2nd ed.

 θ_c . Thus, (4.1) becomes, neglecting, $1/C_l$,

$$\frac{1}{\tau_{sp}} \approx 4 \left(\frac{\hbar}{Mk^2}\right) \left(\frac{\theta_D^2}{\theta_c^4}\right) S^2 [\alpha J(R_h^0)]^2, \qquad (4.4)$$

which is independent of temperature. This point has not been noted before; however, one must bear in mind that the spin-wave approximation breaks down in the high-temperature regions and there may not be any physical significance in calculations pertaining to the high-temperature limit.

Two-Phonon Process

Corresponding to the two cases implied in equations (3.21) and (3.23) the relaxation times for the twophonon processes at low temperatures are given by

$$\left(\frac{1}{\tau_{sp}}\right)_{R_1} \approx \frac{1}{\pi^5} \left(\frac{\hbar^3}{M^2 k^3}\right) \left(\frac{T}{\theta_D}\right)^{10} \left(\frac{T}{\theta_D^3 \theta_c}\right) \left(11 + \frac{\beta^{\frac{3}{2}}}{234}\right) \\ \times S^2 J_R^2 \quad (4.5)$$
and

$$\left(\frac{1}{\tau_{sp}}\right)_{R_2} \approx \frac{1}{\pi^5} \left(\frac{\hbar^3}{M^2 k^3}\right) \left(\frac{T}{\theta_D}\right)^{10} \left(\frac{1}{\theta_c^{2\theta_D}}\right) \left(11 + \frac{\beta^{\frac{3}{4}}}{234}\right) \\ \times S^2 J_R^2. \quad (4.6)$$

The corresponding expressions for the high-temperature limit can be obtained from (3.21) and (3.23) on dividing them by $\Delta TNk/6\pi^2$.

5. ESTIMATES AND DISCUSSION

In the expressions for the various relaxation times derived in the previous section, most of the factors are easily determined except those involving $^{\alpha}J(R_{h}^{0})$ and J_R [cf. (2.13) and (3.22)]. A knowledge of these will require the calculation of the exchange integrals involving excited orbitals as well as the matrix elements of the various derivatives of the crystal field potentials with respect to excited and ground state orbitals. We consider an order-of-magnitude estimate of these quantities for a body-centered cubic system such as iron. The excited orbitals for iron are taken to be the 4p empty orbitals. Thus, the transitions involved are 3d to 4p, which for the cubic symmetry of the systems can be achieved owing to odd vibrations of the surrounding ions with respect to the ion in question. For the actual estimate, we consider a collinear system of three ions lying on the body diagonal of the unit cell. The potential energy of an electron at the central ion in the field of the two diagonally opposite nearest neighbors at the equilibrium position can be taken as¹²

$$V = -\left(\frac{Ze^2}{R_0 - r} + \frac{Ze^2}{R_0 + r}\right),$$
(5.1)

where R_0 is the distance of the neighbor from the central ion, r is the coordinate of the electron referred to the central ion, and +|Ze| is the charge of the ion. For an odd vibration of the unit chosen, the first and second derivatives of the potential are

$$|V^h| = (\partial V/\partial R_h)_0 = 4Ze^2 r/R_0^3, \qquad (5.2)$$

$$V^{hh}| = (\partial^2 V / \partial R_h^2)_0 = 6Z e^2 r / R_0^4.$$
(5.3)

Thus, the matrix elements required have the approximate values

$$|\psi_{3dz^2}| V^h |\psi_{4pz}\rangle \sim 4Z e^2 r_0 / R_0^3,$$
 (5.4)

$$\langle \psi_{3dz^2} | V^{hh} | \psi_{4pz} \rangle \sim 6Z e^2 r_0 / R_0^4,$$
 (5.5)

with r_0 standing for the radius of the ion. For iron the charge of the ion core is taken for the ion without the 4s electrons, i.e., +2|e|, and the radius $r_0 \sim 1$ Å. Thus with $R_0 = 2.5$ Å the value of $\langle \psi_{3dz^2} | V^h | \psi_{4pz} \rangle \sim 1.25$ $\times 10^{-3}$ dyn. Likewise the value of (5.6) is 0.75×10^{5} dyn/cm.

The energy denominator $\Delta E_{\alpha} = E_{\alpha} - E_0$ is taken to be the difference in 4p and 3d bands for iron-series metals. This is estimated from x-ray spectroscopy data¹⁵ as $\Delta E_{\alpha} \approx 5$ eV. A reasonable range will be 5 to 10 eV; however, to be on the safe side we shall use the value $\Delta E_{d-p} \sim 10$ eV. For an estimate of the exchange integrals of the type $\langle \psi_{4p}{}^{(1)}\psi_{3d}{}^{(2)} | g_{12} | \psi_{3d}{}^{(1)}\psi_{3d}{}^{(2)} \rangle$ we make use of the calculations of similar integrals in certain other magnetic studies.^{16,17} The value is of the order of 0.01 atomic unit (0.27 eV). We use the value of 0.1 eV in our estimate.

Thus for iron, at $T = 10^{\circ}$ K, $\theta_c = 1000^{\circ}$ K, and θ_D =500°K, we get the estimate of $(1/\tau_{sp}) \sim 2.5 \times 10^6$ sec⁻¹, i.e.,

$$\tau_{sp} \approx 10^{-6}$$
 sec,

for the one-phonon process with S=1 [cf. Eq. (4.3)]. This estimate seems to be of the right order of magnitude as can be seen by comparing with the values suggested earlier.^{3,5} For the high-temperature limit [cf. Eq. (4.4)], we get the value $(1/\tau_{sp}) = 5 \times 10^{10} \text{ sec}^{-1}$, i.e., the relaxation time τ_{sp} is of the order of 10^{-11} sec. This value may be taken to be the limiting value since $(1/\tau_{sp})$ in this limit becomes independent of temperature. From this it is reasonable to expect that at room temperature τ_{sp} will be around 10^{-8} to 10^{-9} sec in agreement with that suggested by Bloembergen and Damon.18

The estimates of $(1/\tau_{sp})_{R_1}$ and $(1/\tau_{sp})_{R_2}$ [cf. (4.5) and (4.6)] arising out of two-phonon processes, following the above procedure, show that the values at low temperature are extremely small. Hence, these processes are unimportant compared to one-phonon processes

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 ¹⁵ C. Mande, Ann. phys. (Paris) 15, 1559 (1960).
 ¹⁶ S. Koide, K. P. Sinha, and Y. Tanabe, Prog. Theoret. Physics (Kyoto) 22, 647 (1959).
 ¹⁷ R. K. Nesbet, Phys. Rev. 119, 658 (1960).
 ¹⁸ N. Bloembergen and R. W. Damon, Phys. Rev. 85, 699 (1952).

^{(1952).}

discussed above. They may, however, become appreciable at higher temperatures.

The foregoing analysis shows that the mechanism suggested here is of importance in the relaxation processes involving the ordered spin and lattice systems. Although the estimates have been made for iron, the process may have wider validity for other systems, such as the ferrites and the garnets. That such mixing of excited states with ground states owing to certain types of vibrations does occur is vindicated by the absorption spectra study of systems containing the transition metal ions.19

We briefly discuss the mechanism in this paper in relation to those suggested earlier.^{3,7} The coupling terms arising out of the expansion of exchange interaction energy in powers of displacements³ or strain tensors⁷ do not provide an unambiguous atomistic picture of the physical process and the terms are difficult to evaluate from first principles. On the other hand, the interaction terms in the present paper stem from a change in electron-lattice potential energy due to crystal field oscillations which in turn leads to

$$^{\alpha}J(R_{lm}) = \langle \phi_{\alpha}\phi_{m} | g_{12} | \phi_{m}\phi_{l} \rangle \langle \phi_{\alpha} | V^{h} | \phi_{l} \rangle / (E_{\alpha} - E_{l}),$$

and thus determines the strength of the phononmagnon interactions. Such factors occurring in (4.3) can be calculated explicitly in principle. Also, they furnish a clear physical picture of the microscopic processes involved.

Further, the one-phonon process in the phenomeno-

logical theory is written as

$$\lambda_{iklmrs} \int \frac{\partial M_i}{\partial x_k} \frac{\partial M_l}{\partial x_m} u_{rs} dV,$$

where the M_i 'S are the components of the vector density of the magnetic moment of the whole crystal, λ_{iklmrs} is a coupling constant, and $u_{rs} = \frac{1}{2} (\partial u_r / \partial x_s)$ $+\partial u_s/\partial x_r$) is the strain tensor, **u** being the displacement vector. Since such interaction terms may be considered as the expansion of the exchange energy in power of strain tensors, the expression [e.g., Eq. (25) of reference 77 contains an additional wave-vector factor compared to our expression [cf. (3.11)]. Thus, at low temperatures the mechanism considered in the present work would seem to be more effective. A more explicit comparison between the two approaches is, however, not possible in that it does not seem approppriate to express the present mechanism in terms of the phenomenological concepts.

The motivation behind our study is more towards understanding the exchange of energy between the phonon and magnon systems in relation to certain physical properties such as thermal conductivity and absorption of sound rather than to the microwave absorption. The details of the calculations for such studies including the spin-orbit interaction, anharmonic coupling of lattice vibrations, etc., will be considered in another paper.

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Curie Point and Origin of Weak Ferromagnetism in Hematite

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The Curie point of α -Fe₂O₃ is found by differential thermal analysis (DTA) to be at 725°C, rather than the value of 675°C known from magnetic measurements. Of the theories on the origin of the weak ferromagnetism in this material, only that of Dzialoshinskii can account for all the experimental data. The 675°C is argued to be a point at which the basal plane anisotropy (assumed nonsymmetric) changes sign.

HEMATITE, α -Fe₂O₃, is a rhombohedral natural crystal having the symmetry $R\overline{3} 2/c$ with¹ a=5.4243 Å and $\alpha=55^{\circ}17.5'$. It contains four iron ions per unit cell, which are located along the [111] direction. The material is, to a first approximation, antiferromagnetic. Below $\sim 250^{\circ}$ K, the magnetization of the sub-

¹ F. Bertaut, Compt. rend. 246, 3335 (1958).

It is known³ that a weak ferromagnetism is superimposed on the antiferromagnetism of this material and this is most pronounced in the (111) plane, and in the

² G. G. Shull, W. A. Strauser, and E. D. Wollan, Phys. Rev. 83, 333 (1951). ³ L. Néel, Revs. Modern Phys. 25, 58 (1953).

¹⁹ D. S. McClure, *Solid-State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1959), Vol. 9.

lattices is parallel to [111], while above 250°K it is in the (111) plane.²