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Light Scattering by Nuclear Magnons in Ferro - and Anti-Ferromagnets*

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We present a detailed theory of light scattering by nuclear magnons in ferro- and antiferromagnets. Three mechanisms of coupling photons with nuclear spins are analyzed and compared. Calculations are developed for a one-nuclear magnon process and second-order processes involving nuclear and electronic magnons in antiferromagnets. A comparison of the Raman intensities of these processes with the purely electronic ones shows that an experimental investigation is feasible.

Apresenta-se uma teoria detalhada do espalhamento de luz por magnons nucleares em ferro- e antiferromagnetos. Três maneiras de acoplar fotons com spins nucleares são analisadas e comparadas. Apresentam-se, em antiferromagnetos, cálculos para processos envolvendo um magnon nuclear, como também para processos de segunda ordem em que comparecem magnons de origem nuclear e eletrônica. Conclue-se, pela comparação das intensidades Kaman desses processos com aqueles puramente eletrônicos, ser factível uma investigação experimental da situação física discutida.

I. Introduction

Nuclear magnons are well defined collective excitations in magnetic materials. These excitations are due to an indirect coupling between nuclear and electronic spins and have frequencies in the nuclear magnetic resonance (NMR) range. This indirect coupling was first proposed by Suhl[1] and Nakamura[2] to explain the linewidth of the NMR of non-magnetic ions in ordered materials. De Gennes et al. later showed that this coupling leads to a k-dependent pulling of the nuclear frequency and that the dispersion relation becomes approximately $\omega_k = \omega_N(1 - y\,\mathcal{H}_N/\Omega_k)$ for a ferromagnet and, for a two-sublattice antiferromagnet, $\omega_k = \omega_N(1 - 2\gamma^2\,\mathcal{H}_E\,\mathcal{H}_N/\Omega_{1k}\Omega_{2k})^{1/2}$, in the normal state, and $\omega_{ik} = \omega_N(1 - 2\gamma^2\,\mathcal{H}_E\,\mathcal{H}_N/\Omega_{ik}^2)$ for the i^{th} mode in the flopped state. In the above, γ is the electronic gyromagnetic ratio, ω_N is the unpulled

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NMR frequency, Ω_k is the electronic spin wave frequency and, $\mathscr{H}_E$ and $\mathscr{H}_N$, are exchange and hyperfine fields, respectively.

The percentage of pulling for a ferromagnet, $y\,\mathscr{H}_N/\Omega_k$, is not significant under typical conditions, since $\mathscr{H}_N$ is about 2 Oe at 4.2°K, and Ω_0/γ is usually of 1 000 Oe. In a low-anisotropy ferromagnet, however, the pulling, $2\gamma^2\mathscr{H}_E\mathscr{H}_N/\Omega_{ik}^2$ or $2\gamma^2\mathscr{H}_E\mathscr{H}_N/\Omega_{1k}\Omega_{2k}$, can be comparable to unity since we then have $\gamma H, \gg \Omega_k$ at low k.

Conventional NMR measurements give information only about the nuclear uniform precession (k = 0). In recent years, microwave photon[4-6] and phonon[7,8] non-linear pumping techniques have been used to probe the excitation of nuclear magnons. These techniques lead only to indirect information about the nuclear modes and have several limitations[4-10]. In this work, we show that the technique of inelastic light scattering can also be used to study nuclear spin waves. Three mechanisms can be envisaged which allow the coupling of light photons with nuclear magnons, namely: **a.** Direct magnetic-dipole interaction analogous to the case of electronic magnons'[1]; L. Indirect coupling of the nuclear spins with the electric field of the radiation via the electric-dipole interaction with the electrons combined with the hyperfine interaction; **c.** Indirect coupling via virtual electronic magnons. We show that, in usual situations, mechanism **c.** is much stronger than the others.

In Sec. 2, we review some results[5] from the theory of nuclear magnons in ferro- and antiferromagnets which will be needed later. In Sec. **3,** we outline the calculations of interactions a., b. and **c.** .

2. Nuclear Magnons

A. Ferromagnets

Let us consider a system with the following Harniltonian

$$H = -\sum_{ij} J_{ij}\,\mathbf{S}_i \cdot \mathbf{S}_j - \gamma_e \hbar\,\mathscr{H}_0 \sum_i S_i^z - \gamma_N \hbar\,\mathscr{H}_0 \sum_i I_i^z + A \sum_i \mathbf{I}_i \cdot \mathbf{S}_i, \qquad (1)$$

where the terms, in order, represent the exchange energy, the electronic and nuclear Zeeman energies, and the electron-nucleus hyperfine coupling. The magnetic field H is in the *z*-positive direction, and γ_e, γ_N are respectively the electronic and nuclear magnetogyric ratio.

The nuclear and electronic spectra will be derived using the Holstein-Primakoff method in the linear approximation, i.e.,

$$\begin{aligned} S^+ &= (2S)^{1/2}a_i^\dagger, \qquad S_i^- = (2S)^{1/2}a_i, \qquad S_i^z = -S + a_i^\dagger a_i, \\ \mathbf{1} &= (2\langle I^z\rangle)^{1/2}b_i, \quad \mathbf{1} = (2\langle I^z\rangle)^{1/2}b_i^\dagger, \quad I_i^z = \langle I^z\rangle - b_i^\dagger b_i, \end{aligned} \tag{2}$$

where $[a_i, a_i^\dagger] = [b_i, b_i^\dagger] = 1$, for all i, all other commutators vanishing. The linearization implied by the above use of $\langle I^z\rangle$ has been justified in detail by De Gennes et *al.*[3].

Substitution of Eq. (2) in Eq. (1), with neglect of quartic terms, followed by the canonical transformations

$$a_k = N^{-1/2}\sum_j \exp(-\mathrm{i}k.\mathbf{R}_j)a_j, \quad b_k = N^{-1/2}\sum_j \exp(\mathrm{i}k.\mathbf{R}_j)b_j, \tag{3}$$

where N is the number of unit cells in the crystal, yields

$$H = c + \sum_k [A_k a_k^\dagger a_k + B\, b_k^\dagger b_k + F(a_k b_k + a_k^\dagger b_k^\dagger)]. \tag{4}$$

where

$$\begin{aligned} A_k &= -\gamma_e h_0 M + A\langle I^z\rangle + J_0 - J_k, \\ B &= \gamma_e h_0 M + AS, \quad F = A(\langle I^z\rangle S)^{1/2}. \end{aligned} \tag{5}$$

and

$$J_k = 2S\sum_{i,j} J_{ij}\exp[\mathrm{i}k.(\mathbf{R}_i - \mathbf{R}_j)].$$

The Hamiltonian H is diagonalized via the canonical transformation

$$\begin{aligned} a_k &= \alpha_k \cosh\theta_k + \beta_k^\dagger \sinh\theta_k, \\ b_k &= \alpha_k^\dagger \sinh\theta_k + \beta_k \cosh\theta_k, \end{aligned} \tag{7}$$

where

$$\tanh 2\theta_k = -2F/(A_k + B). \tag{8}$$

There results

$$H = c - \frac{1}{2}\sum_k (A_k + B) + \sum_k (\varepsilon_{k\alpha}\alpha_k^\dagger\alpha_k + \varepsilon_{k\beta}\beta_k^\dagger\beta_k), \tag{9}$$

where

$$\begin{aligned} \varepsilon_{k\alpha} &= \frac{1}{2}[(A_k - B) + (A_k + B)/\cosh 2\theta_k], \\ \varepsilon_{k\beta} &= \frac{1}{2}[-(A_k - B) + (A_k + B)/\cosh 2\theta_k]. \end{aligned} \tag{9}$$

Since ω_N, the unpulled frequency, is equal to $AS/\hbar$ and $\mathscr{H}_N$, the field seen by the electrons due to the nucleus, is equal to $A\langle I^z\rangle/\gamma_e\hbar$, we have for typical values $|2F/(A_k + B)| \ll 1$, and

$$\begin{aligned}
&(\cos\ 2\theta_k)^{-1} \simeq 1 - \frac{1}{2}\tanh^2\ 2\theta_k = 1 - 2F^2/(A_k + B)^2,\\
&\varepsilon_{k\alpha} \simeq A_k - F^2/(A_k + B) \simeq \hbar\Omega_k - \hbar\gamma_e\mathscr{H}_N\omega_N/\Omega_k,\\
&\varepsilon_{k\beta} \simeq B - F^2/(A_k + B) \simeq \hbar\omega_N(1 - \gamma_e\mathscr{H}_N/\Omega_k),
\end{aligned} \tag{10}$$

where Ω_k is the electronic magnon frequency. Thus, from Eq. (10), we see that α and β are, respectively, the electronic and nuclear spin-wave modes.

B. Antiferromagnets

We now consider the cubic antiferromagnet in the unflopped state (to the flopped state see Ref. 13). The Hamiltonian, we consider now, is

$$\begin{aligned}
H = {} & 2J\sum_{ij}\mathbf{S}_i\cdot\mathbf{S}_j + \gamma_e\hbar(\mathscr{H}_A - \mathscr{H}_0)\sum_i S_i^z - \gamma_E\hbar(\mathscr{H}_A + \mathscr{H}_0)\sum_j S_j^z\\
& + \gamma_N\hbar\mathscr{H}_0\Big[\sum_i I_i^z + \sum_j I_j^z\Big] - A\Big[\sum_i \mathbf{I}_i\cdot\mathbf{S}_i + \sum_j \mathbf{I}_j\cdot\mathbf{S}_j\Big],
\end{aligned} \tag{11}$$

where i and j refer to A and B sublattices, respectively. Each term, in the above, means: antiferromagnetic exchange ($J > 0$); combined electronic Zeeman and anisotropy energy for the *A* sublattice and for the B sublattice; nuclear Zeeman energy and hyperfine interaction.

From usual spin-wave theory, we set

$$\begin{aligned}
&S_i^+ = (2S)^{1/2}a_i, \quad S_i^- = (2S)^{1/2}a_i^\dagger, \quad S_i^z = S - a_i^\dagger a_i,\\
&S_j^+ = (2S)^{1/2}b_j^\dagger, \quad S_j^- = (2S)^{1/2}b_j, \quad S_j^z = -S + b_j^\dagger b_j,\\
&I_i^+ = (2\langle I_A^z\rangle)^{1/2}c_i, \quad I_i^- = (2\langle I_A^z\rangle)^{1/2}c_i^\dagger, \quad I_i^z = \langle I_A^z\rangle - c_i^\dagger c_i,\\
&I_j^+ = (2\langle I_B^z\rangle)^{1/2}d_j^\dagger, \quad I_j^- = (2\langle I_B^z\rangle)^{1/2}d_j, \quad I_j^z = -\langle I_B^z\rangle + d_j^\dagger d_j,
\end{aligned} \tag{12}$$

where $[a_i, a_i^\dagger] = [b_j, b_j^\dagger] = [c_i, c_i^\dagger] = [d_j, d_j^\dagger] = 1$, all other pairs of operators commuting.

Substitution of Eq. (12) in Eq. (11), followed by the canonical transformations

$$\begin{aligned}
&a_k = N^{-1/2}\sum_i \exp(i\mathbf{k}\cdot\mathbf{R}_i)a_i, \quad b_k = N^{-1/2}\sum_j \exp(-i\mathbf{k}\cdot\mathbf{R}_j)b_j,\\
&c_k = N^{-1/2}\sum_i \exp(i\mathbf{k}\cdot\mathbf{R}_i)c_i, \quad d_k = N^{-1/2}\sum_j \exp(-i\mathbf{k}\cdot\mathbf{R}_j)d_j,
\end{aligned} \tag{13}$$

where N is the number of unit cells in the crystal, leads to

$$H = \hbar \sum_{k} [\tilde{A} a_k^{\dagger} a_k + D_1 c_k^{\dagger} c_k - F_A(a_k^{\dagger} c_k + a_k c_k^{\dagger}) + B b_k^{\dagger} b_k + D_2 d_k^{\dagger} d_k - F_B(b_k^{\dagger} d_k + b_k d_k^{\dagger}) + \omega_{ex}\gamma_k(a_k b_k + a_k^{\dagger} b_k^{\dagger})]. \quad (14)$$

The coefficients, in Eq. (14), are

$$\begin{aligned} \tilde{A} &= \omega_{ex} - \gamma_e(\mathcal{H}_A - \mathcal{H}_0 + \mathcal{H}_{NA}), \quad \mathcal{H}_{NA} = -A\langle I_A^z\rangle/\gamma_e\hbar, \\ B &= \omega_{ex} - \gamma_e(\mathcal{H}_A + \mathcal{H}_0 + \mathcal{H}_{NB}), \quad \mathcal{H}_{NB} = -A\langle I_B^z\rangle/\gamma_e\hbar, \\ \omega_{ex} &= 2SzJ/\hbar, \quad D_1 = \omega_N + \gamma_N\mathcal{H}_0, \quad \omega_N = AS/\hbar, \\ D_2 &= \omega_N - \gamma_N\mathcal{H}_0, \; F_A = (-\gamma_e\omega_N\mathcal{H}_{NA})^{1/2}, \; F_B = (-\gamma_e\omega_N\mathcal{H}_{NB})^{1/2}, \end{aligned} \quad (15)$$

the last sum being over the z nearest neighbors of a given site.

The Hamiltonian in Eq. ***(14)*** can be diagonalized as follows. First, we express H in matrix form as

$$H = X^{\dagger} H X, \quad (16)$$

where

$$X = \begin{pmatrix} a_k \\ c_k \\ b_k^{\dagger} \\ c_k^{\dagger} \end{pmatrix}, \quad H = \begin{pmatrix} \tilde{A} & -F_A & \gamma_k\omega_{ex} & 0 \\ -F_A & D_1 & 0 & 0 \\ \gamma_k\omega_{ex} & 0 & B & -F_B \\ 0 & 0 & -F_B & D_2 \end{pmatrix} \quad (17)$$

Knowing that the elements of X are boson operators allows us to write the following matrix equation:

$$g\,H\,S = S\,g\,\Omega, \quad (18)$$

where

$$g = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix}, \quad \Omega = \begin{pmatrix} \omega_1 & 0 & 0 & 0 \\ 0 & \omega_2 & 0 & 0 \\ 0 & 0 & \omega_3 & 0 \\ 0 & 0 & 0 & \omega_4 \end{pmatrix} \quad (19)$$

and S is the transformation matrix which diagonalizes H and defines the normal mode operators by

$$Y = SX, \quad (20)$$

where

$$Y = \begin{pmatrix} \alpha_{k_1} \\ \alpha_{k_2} \\ \alpha_{k_3} \\ \alpha_{k_4} \end{pmatrix} \tag{21}$$

The eigenvalues are given by

$$[(A-\omega)(B+\omega) - \gamma_k^2\omega_{ex}^2](D_1-\omega)(D_2+\omega) - F_A^2(B+\omega)(D_2+\omega) - \\ - F_B^2(\tilde{A}-\omega)(D_1-\omega) + F_A^2F_B^2 = 0. \tag{22}$$

The roots of the above equation, for a low-anisotropy antiferromagnet, are given approximately by

$$\begin{aligned} \omega_1 &\simeq x_1 - \gamma_e\omega_N\mathscr{H}_N(x_1^2 + \omega_{ex}\omega_N)/x_1^3, \\ \omega_2 &\simeq D_1 - \omega_N[1 - (1 + 2\gamma_e\mathscr{H}_N\omega_{ex}/x_1\,x_2)^{1/2}], \\ \omega_3 &\simeq -x_2 + \gamma_e\omega_N\mathscr{H}_N(x_2^2 + \omega_{ex}\omega_N)/x_2^3, \\ \omega_4 &\simeq -D_2 + \omega_N[1 - (1 + 2\gamma_e\mathscr{H}_N\omega_{ex}/x_1\,x_2)^{1/2}], \end{aligned} \tag{23}$$

where

$$x_1 = \left[\frac{1}{4}(\tilde{A}-B)^2 + \tilde{A}B - \omega_{ex}^2\gamma_k^2\right]^{1/2} + \frac{1}{2}(\tilde{A}-B),$$

and

$$x_2 = \left[\frac{1}{4}(\tilde{A}-B)^2 + \tilde{A}B - \omega_{ex}^2\gamma_k^2\right]^{1/2} - \frac{1}{2}(\tilde{A}-B), \tag{24}$$

are the unperturbed electronic frequencies. From Eqs. (23), we see that $\omega_{1,3}$ and $\omega_{2,4}$ are the frequencies of the quasi-electronic and nuclear modes respectively. In Fig. 1, we sketch the k-dependence for these frequencies.

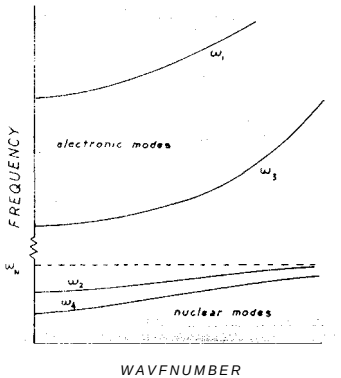

Fig. 1 – Dispersion relation for coupled nuclear and electronic spin wave modes in a two-sublattice antiferromagnet. Typically, the curves bend up at k ~ 10^5 cm^{-1}

Performing the algebra, we have for S, in Eq. (18),

$$S = \begin{vmatrix} \dfrac{\gamma_k\omega_{ex}(D_1-\omega_1)\mu_1}{(\tilde{A}-\omega_1)(D_1-\omega_1)-F_A^2} & \dfrac{\gamma_k\omega_{ex}F_A\mu_1}{(\tilde{A}-\omega_1)(D_1-\omega_1)-F_A^2} & \mu_1 & \dfrac{F_B\mu_1}{D_2+\omega_1} \\ \mu_2 & \dfrac{F_A\mu_2}{D_1-\omega_2} & \dfrac{\gamma_k\omega_{ex}(D_2+\omega_2)\mu_2}{(B-\omega_2)(D_2+\omega_2)-F_B^2} & \dfrac{\gamma_k\omega_{ex}F_B\mu_2}{(B+\omega_2)(D_2+\omega_2)-F_B^2} \\ \mu_3 & \dfrac{F_A\mu_3}{D_1-\omega_3} & \dfrac{\gamma_k\omega_{ex}(D_2+\omega_3)\mu_3}{(B+\omega_3)(D_2+\omega_3)-F_B^2} & \dfrac{\gamma_k\omega_{ex}F_B\mu_3}{(B+\omega_3)(D_2+\omega_3)-F_B^2} \\ \dfrac{\gamma_k\omega_{ex}(D_1-\omega_4)\mu_4}{(\tilde{A}-\omega_4)(D_1-\omega_4)-F_A^2} & \dfrac{\gamma_k\omega_{ex}F_A\mu_4}{(\tilde{A}-\omega_4)(D_1-\omega_4)-F_A^2} & \mu_4 & \dfrac{F_B\mu_4}{D_2+\omega_4} \end{vmatrix} \tag{25}$$

where the μ's satisfy

$$|\mu_j|^2 \left\{1 + \frac{F_A^2}{(D_1-\omega_j)^2} - \frac{\gamma_k^2\omega_{ex}^2[(D_2+\omega_j)^2+F_B^2]}{[(B+\omega_j)(D_2+\omega_j)-F_B^2]^2}\right\} = \begin{matrix} 1, & j=2, \\ -1, & j=3, \end{matrix}$$

$$|\mu_j|^2 \left\{\frac{\gamma_k^2\omega_{ex}^2[(D_1-\omega_j)^2+F_A^2]}{[(\tilde{A}-\omega_j)(D_1-\omega_j)-F_A^2]^2} - 1 - \frac{F_B^2}{(D_2+\omega_j)^2}\right\} = \begin{matrix} 1, & j=1, \\ -1, & j=4. \end{matrix} \tag{26}$$

3. Light Scattering

Mechanism a. is analogous to the process suggested by Bass and Kaganov[11] for Kaman scattering by electronic magnons. Fleury and Loudon[12] have shown that, in the electronic magnon case, this interaction is negligible compared to the indirect electric-dipole process. Their arguments also apply here and we shall not consider this process further. To calculate interactions b. and c., we use the single ion model used by Fleury and Loudon[12] to study light scattering by electronic magnons, except that we allow for the presence of the hyperfine splitting in the energy levels. Consider a crystal in which the ground state of the magnetic ion has spin S, zero orbital angular momentum ($L=0$) and nuclear spin I. The intermediate states, connected to the ground state by the electric dipole moment, must have $L=1$ and the same electronic and nuclear spins, S and I. The ground state is split in $(2S+1)(2I+1)$ components by the external field, the exchange field and the hyperfine interaction. The energy diagram is sketched in Fig. 2. We assume that the splitting between components of different S_z (which corresponds approximately to the electronic magnon energy) is much larger than the splitting between components with the same S_z and different I_z (approximately, the nuclear magnon energy). In the excited

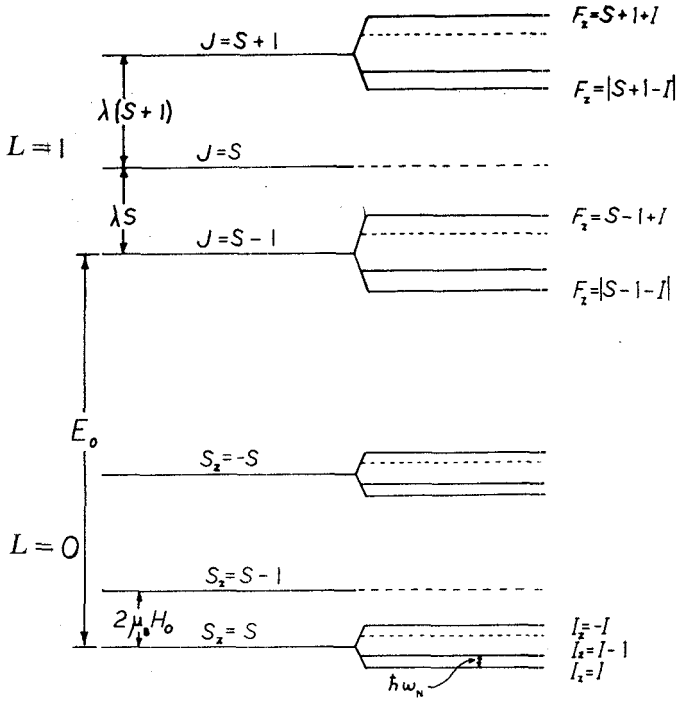

Fig. 2 – Energy level diagram used to calculate the Rarnan processes for nuclear and electronic spins.

P-state, we neglect the splitting in S, in comparison with the spin-orbit coupling splitting. In the excited state, each of the three J components splits in F-components due to the hyperfine interaction. Here, $\mathbf{F} = \mathbf{J} + \mathbf{I}$ follows the angular momenta sum rules. The energy eigenfunctions $|F\ F,)$ for the excited states can be expressed as linear combinations of $|J_z\ \mathbf{I},)$ eigenfunctions, which are in turn linear combinations of $(L,S,\ I,)$ eigenfunctions. The electric-dipole interaction between the radiation and the electrons of the magnetic ions ($\mathscr{H}_{\mathrm{ED}} = -\ \mathrm{e} \sum_i \mathbf{E}_i \,.\, \mathbf{r}_i$)

connects an initial state $S_z^i\ I_z^i$ with intermediate states and these with a final state $S_z^f\ I_z^f$. S pin-orbit and hyperfine interactions mix different S, and I, in the intermediate states, allowing the flip of electronic and nuclear spins. One can, therefore, calculate the tsansition probabilities for different Raman processes, involving changes in I, and/or S_z. With $I_z^f = I_z^i$ and $S_z^f = S_z^i \pm 1$, the calculation is identical to that of Fleury

and Loudon[12]. With third-order perturbation theory, one can represent the Raman process in terms of an electronic spin operator Hamiltonian[12] given by

$$H_{s-\mathrm{rad}} = \Gamma_s \sum_i (E_L^z E_S^+ - E_L^+ E_S^z) S_i^- + \mathrm{h.c.} \tag{27}$$

where

$$\Gamma_s = (e^2\lambda/2^{3/2}) \langle 00 | z | 10 \rangle \langle 1 - 1 | r^- | 00 \rangle \times \\ \times \{[1/(E_0 - \hbar\omega_L)^2] - [1/(E_0 + \hbar\omega_s)^2]\}, \tag{28}$$

the summation running over all magnetic sites i, $r^- = x - iy$; E_0 is the energy separation between the ground state and the lowest allowed intermediate state, λ the spin-orbit coupling parameter of the excited state, E^z and $E^+ = E^x + i E^y$ the electric field components of the radiation with frequency ω and the subscripts L and S refer to the incident and scattered fields, respectively. In the matrix elements, the kets $|L L,)$ characterize the orbital part of the ground and excited states. Now, setting $S_z^f = S_z^i$ and $I_z^f = I_z^i \pm 1$ and following the same steps which led to Eq. (27), we find the Hamiltonian for the interaction between the radiation fields and the nuclear spins

$$H_{I-\mathrm{rad}} = \Gamma_I \sum_i (E_L^z E_S^+ - E_L^+ E_S^z) I_i^- + \mathrm{h.c.} \tag{29}$$

where

$$\Gamma_I = \frac{A_L}{\lambda} \Gamma_s, \tag{30}$$

where A_L is the orbital hyperfine constant of the excited states. In Eq. (30), we have neglected λ in comparison with E_0. The Hamiltonian (29) represents a process analogous to the one of (27), in which the spin-orbit interaction allows the flipping of the electronic spin by the electric field of the radiation. Here, the hyperfine coupling needed to flip the nuclear spins proceeds through the orbital momentum of excited non-S states of the magnetic ions which serve as intermediate states in the Raman process.

Mechanism b., referred to previously, arises from the interaction Hamiltonian (29). In ferromagnets, the nuclear spin deviation operators I_i^- and I_i^+ can be expressed directly in terms of nuclear magnon creation and destruction operators. Therefore, (29) leads to Stokes and anti--Stokes scatterings processes very much analogous to the electronic

case[12]. Due to relation (30), the intensity of this scattering process for nuclear magnons is smaller than for electronic magnons by a factor of $(A_L/\lambda)^2$. For Mn ions this ratio is of the order of 10^{-10} and mechanism b. is too weak.

Finally, mechanism *c.* arises from the interaction Hamiltonian (27). With the hyperfine coupling present, an electron deviation is necessarily accompanied by a disturbance of the nuclear systems, and this is seen in Eq. (7), the transformation that diagonalizes the Hamiltonian of a spin system with electrons and nuclei coiipled by the hyperiine interaction. Replacing the spin operator in Eq. (27) by the second relation in Eq. (2) and using **Eq. (7)**, we obtain the interaction Hamiltonian for the first-order Stokes scattering process by nuclear magnons,

$$H^{s}_{N-rad} = [(2\pi\hbar)(2\omega_L\omega_S SN)^{1/2}/\eta_L\eta_S v]\,\Gamma_s \sinh\theta_k \times \times \sum_{k_N} (\varepsilon^z_L\varepsilon^+_S - \varepsilon^+_L\varepsilon^z_S)\, a_L a^{\dagger}_S \beta^{\dagger}_k\, \delta(\mathbf{k}_L - \mathbf{k}_S - \mathbf{k}_N) + \text{h.c.}, \quad (31)$$

where

$$\sinh\theta_k \simeq -(\gamma\mathscr{H}_N\omega_N)^{1/2}/(\Omega_k + \omega_N),$$

$\eta_{L,S}$ and $\varepsilon_{L,S}$ are, respectively, the refractive indices and the polarizations of the incident and scattered radiations, a_L and $a^{\dagger}_S$ their photon destruction and creation operators and, v, the interaction volume.

Comparison between (31) and (27) shows that, in ferromagnets, the intensity of the scattering by nuclear magnons in the present process is smaller than by electronic magnons by a factor $\sinh^2\theta_k$. In typical ferromagnets, this factor can be as large as 10^{-3} and mechanism c. is much stronger than the others. The polarization selection rules, for scattering by nuclear magnons, are the same as those for electronic magnons[12].

We can now use mechanism c. to obtain the Hamiltonian for scattering in antiferromagnets by allowing the summation in (27) to run over the different magnetic sublattices and by use of the appropriate transformations from the spin to the normal mode operators of Eqs. (12) and (20). With this Hamiltonian, we arrive at the differential cross-section for Stokes scattering by the i^{th} -mode nuclear spin wave in a two-sublattice unflopped antiferromagnet,

$$\frac{d\sigma}{d\Omega} = [2M v\,\eta_S\omega_L\omega_S^3(n^i_k + 1)\Gamma_s^2(S_{pi} + S_{qi})^2/g\mu_B\eta_L c^4]\,|\varepsilon^z_L\varepsilon^-_S - \varepsilon^-_L\varepsilon^z_S|^2, \quad (32)$$

where, M, is the sublattice magnetization, $g\mu_B$ is the elementary electronic magnetic moment and, n_k^i, the i^{th} -mode nuclear magnon ocuppation number. S_{pi} and S_{qi} are the matrix elements from Eq. (25) corresponding to the i^{th} -mode nuclear magnon creation operator. Using the results from Eq. (25) in an unflopped antiferromagnet, the scattering cross-section (32) is shown to be proportional to $\Gamma_s^2(S_{41} - S_{43})^2$. With the approximations $\mathcal{H}_N \ll \mathcal{H}_0$ and $\mathcal{H}_A \ll \mathcal{H}_E$ where $\mathcal{H}_0$ is the applied field, this factor reduces to

$$V_N = \Gamma_s^2 \omega_N \delta_{AF}^2 (\mathcal{H}_0 - \mathcal{H}_A)^2 / \gamma_e \mathcal{H}_N (\mathcal{H}_E + \mathcal{H}_0 - \mathcal{H}_A)^2, \qquad (33)$$

where $\delta_{AF} = \gamma_e^2 h_E h_N / \Omega_{1k} \Omega_{2k}$ is the frequency pulling. This is to be compared with the factor for scattering by electronic magnons[12] $Ve = (\mathcal{H}_A / 2\mathcal{H}_E)^{1/2} \Gamma_s^2$. For the low-anisotropy antiferromagnet $RbMnF_3$ ($\mathcal{H}_A = 4.5$ *Oe* at T= 4.2°K), in which nuclear magnons were first observed, with $\mathcal{H}_0 = 2$ *KOe*, at T= 4.2°K ($\delta_{AF} = 0.14$), one has $V_N \sim 10^{-5} \Gamma_s^2 \sim 10^{-2}$ *Ve*.

In addition to one-nuclear magnon scattering, it is possible to have, in antiferromagnets, second-order Raman scattering involving the creation or destruction of a pair of nuclear magnons or a mixed electronic nuclear magnon pair. As in the one-nuclear magnon case, the origin of the mechanism here is based on the coupling of light photons with the electronic spins. The interaction of light with two electronic spins in a antiferromagnet, in a Raman process, can be written as[13]

$$\sum_{\substack{i\,d \\ \alpha\beta\gamma\delta}} E_L^\alpha E_S^\beta B(d) S_i^\gamma S_{i+d}^\delta \qquad (34)$$

where d indicates the neighbors to site *i*. The origin of interaction (34) resides on a coupling through virtual phonons or on an exchange mechanism via electronic excitations[12]. The form of the B tensor can be determined from the symmetry of the magnetic crystal. In $RbMnF_3$ (Ref. 13), the interaction is proportional to $\mathbf{S}_i \cdot \mathbf{S}_{i+d}$ and the largest contributions come from the terms $S_i^+ S_{i+d}^-$ and $S_i^- S_{i+d}^+$, where *i* and i + d refer to the down-spin and up-spin sublattices. With the results from Sec. 2, we can replace the electronic spin operators in these terms by the electron-nuclei normal mode operators. The resulting expression contains, in addition to terms which give the scattering by electronic magnons of different branches[12], terms with creation and destruction operators for pairs of nuclear magnons of different modes and mixed electronic-nuclear magnon pairs. The ratio of the interactions for Stokes scattering, by two-nuclear magnons, to Stokes

scattering, by two-electronic magnons, becomes

$$(S_{21}S_{43} + S_{23}S_{41})/(S_{11}S_{33} + S_{31}S_{13}),$$

which, with the approximations appropriate for $RbMnF_3$, reduces to

$$\omega_N\delta_{AF}^2(1 + \gamma_k^2)(u_k^2 + v_k^2)[\gamma_e h_N + (1 - \gamma_k^2)\omega_N\delta_{AF}^2]^{-1}, \qquad (35)$$

where γ_k, u_k and v_k are the usual coefficients used in the transformation which diagonalizes the electronic spin Hamiltonian in a antiferromagnet[12]. At low k, $\gamma_k \simeq 1$, and the ratio *(35)* is of the order of unity. Therefore, in this region, the scattering by nuclear-magnon pairs is comparable to the scattering by electronic-magnon pairs. The spontaneous scattering by nuclear pairs, however, is expected to be much smaller than the electronic analog. The reason is that, since the wave vectors of the light photons are very small compared with the Brillouin--zone edge value of k, the wave vectors of the magnons excited are nearly equal and opposite and can assume any value. As the density of states increases rapidly with k, the larger contribution to the scattering comes from the neighborhood of the Brillouin-zone edge. Here, as k_N increases, the electronic frequency Ω_k increases rapidly and the admixture of the nuclear-electronic spin wave modes, expressed by the frequency pulling δ_{AF}, vanishes quickly. At the adge of the Brillouin-zone, $\delta_{AF} \simeq h_N/h_E \simeq 10^{-6}$, and the ratio (35) is of the order 10^{-10}.

4. Conclusion

In this paper, we show theoretically the possibility of studying nuclear magnons directly with inelastic light scattering techniques. It can be shown that, for $RbMnF_3$, the ratio (33) is of the order 10^{-2} Ve and this intensity is very low. Together with the small frequency shift of the Brillouin signal (the unpulled NMR frequency of Mn^{55} is of the order 680 GHz), this should make the scattering by thermal nuclear magnons very difficult to observe. However, one has the possibility of increasing the population of nuclear magnons by many orders of magnitude with microwave pumping[4-10] leading to strong signals which may be resolved with the high-resolution experimental methods recently developed[14].

A magnetic system, in which first-order light scattering by nuclear magnons may prove to be more useful, is the uniaxial antiferromagnet MnF_2. In this material, the transverse anisotropy is low enough that the application of a high magnetic field brings the downgoing electronic

magnon branch to the low microwave range[15]. Therefore, with a field just above the spin-flop value which is 93 KOe at $T = 4.2°K$, both electronic frequencies are low and coupling with nuclear magnons is strong. Under these conditions, calculations similar to (33) give $V_N \sim 8 \times 10^{-3} \Gamma_s^2 \sim 10^{-1}$Ve. With this larger intensity, we can possibly use light scattering to study nuclear magnons in MnF_2.

Another possible application is in the study of nuclear magnons in spiral and conical spin structures, such as found in certain rare-earth metals[16]. In these materials, the coupling between nuclear and electronic modes is strong and, therefore, the scattering cross-section is large. In addition; with the very large hyperfine fields found, the NMR frequency is large (e.g., $\omega_N \simeq 6.5$ GHz in holmium) and the resolution is no longer critical for the experiments. Again high-resolution spectroscopy[14] may circumvent the problem created by the fact that one has to scatter light off the surface of the rare-earth metal.

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