Aust. J. Phys., 1974, **27**, 457–70

Phenomenological Theory of Magnon Sideband Shapes in a Ferromagnet with Impurities

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Abstract

A simple model Hamiltonian is used to calculate exactly the line shapes of magnon sidebands in excitonic spectra in a ferromagnet, using Green functions. The effect of a spin defect in the crystal is included in the calculation. In contrast to previous work, no approximations involving Green function decoupling are necessary for this Hamiltonian which, however, contains the essential physical features of the problem. Some one-dimensional crystal line shapes are calculated to illustrate the features of the theory, and application to realistic crystals is discussed.

Introduction

Magnon sidebands, the result of interactions between magnons and excitons, have been observed in the excitonic spectra of magnetic crystals by several experimenters (Green *et al.* 1965; Stevenson 1966; Johnson *et al.* 1966). Tanabe *et al.* (1965), Parkinson and Loudon (1968), Loudon (1968), Parkinson (1969) and Bhandari and Falicov (1972) have put forward theories to explain the magnon sideband based on the mechanism of magnon–exciton interaction proposed by Sugano and Tanabe (1963). All these theories required approximations to obtain results because their Hamiltonians, although physically realistic, were nonlinear, and could not be diagonalized. The Hamiltonian chosen in the present paper retains the physical significance of earlier theories but also includes the effect of an impurity. Computation of the effect of the impurity is made manageable by defining a Hamiltonian which is quadratic in form and is exactly soluble despite the lack of translational invariance caused by the defect. We are able to make a clear physical interpretation of the results. The theory may also be extended to antiferromagnetic crystals near the ground state.

Model Hamiltonian

The phenomenological Hamiltonian is chosen to represent the magnons and excitons (of the Frenkel type) in the crystal, and includes a magnon–exciton interaction term. The defect is accounted for by an interaction term between magnons. The advantage of this Hamiltonian is that it is exactly soluble and one can keep track of the physical significance of the results, in contrast to previous theories where the approximations necessary for solution of the more accurate Hamiltonians have tended to make interpretations more difficult. The present Hamiltonian takes the form

$$\mathscr{H} = \sum_{k} \varepsilon_1(\boldsymbol{k})\, a_k^{+}\, a_k + \sum_{k} \varepsilon_2\, b_k^{+}\, b_k + g \sum_{k} (a_k^{+}\, b_k + b_k^{+}\, a_k) + \gamma N^{-1} \sum_{k,k'} a_k^{+}\, a_{k'}, \qquad (1)$$

for N atoms in the crystal. The operators a_k^+, a_k and b_k^+, b_k are creation and annihilation operators for the magnons and excitons respectively, while $\varepsilon_1(\boldsymbol{k})$ is the magnon energy and ε_2 the exciton energy. We neglect any dispersion of the exciton branch which is generally small. The strength of the magnon–exciton interaction is taken to be independent of wave number. The effect of a spin impurity is given by the last term of equation (1). In general γ would be dependent on $\boldsymbol{k}$ and $\boldsymbol{k}'$, but for simplicity we ignore this here, although it could be treated, in principle at least, under this model. Callaway (1963) has given an expression for $\gamma(\boldsymbol{k}, \boldsymbol{k}')$ (his equation (A3)).

The magnon energy may be written as

$$\varepsilon_1(\boldsymbol{k}) = 2JS \sum_{\Delta} (1 - \cos \boldsymbol{k}.\Delta) - 2zJS^2\rho, \tag{2}$$

where we have set the zero of the energy scale at the ground state energy $-JNSz^2$ and

$$\rho = (J'S' - JS)/JS,$$

with J and S the exchange coupling and spin of the host atoms and J' and S' the corresponding characteristics of the defect. The sum over Δ in equation (2) is over nearest-neighbour sites in the crystal, z being the number of nearest neighbours.

The form of the magnon–exciton coupling proposed by Sugano and Tanabe (1963) is an interaction arising out of the coupling between a pair of nearest-neighbour ions. It is an exchange coupling where one of the ions is raised to an excited electronic state and has its spin component changed by unity, while the other ion has an accompanying unit change in spin to conserve the total spin of the system. Parkinson and Loudon (1968) have used this type of interaction to describe the isotropic magnon–exciton interaction, as well as including it in the electric dipole moment which couples to an external electromagnetic field. Instead of including the interaction at both places in the present Hamiltonian, we have assumed that the external electromagnetic field $E \cos \omega_0 t$ couples only to the excitons. The interaction term chosen represents the exciton interaction of Sugano and Tanabe (1963) in a phenomenological manner.

Calculations

We are interested in the optical absorption of a crystal with the Hamiltonian (1) which is perturbed by a Hamiltonian of the form

$$\mathscr{H} = -\sum_n \boldsymbol{p}_n . \boldsymbol{E}, \tag{3}$$

for an electric dipole moment $\boldsymbol{p}_n$ of the nth ion. We ignore the effect of the magnetic component of the external field and represent the Frenkel exciton as a simple harmonic oscillator. Then the Hamiltonian (3) becomes, in second quantized form,

$$\mathscr{H}_1 = \mathrm{i}\alpha N^{-\frac{1}{2}} \sum_k (b_k^+ - b_k) E \cos \omega_0 t, \tag{4}$$

where the electric field is in the direction of polarization and α represents unit polarization of the ion.

The optical absorption is given by the imaginary part of the Green function

$$G(t - t') = \langle\langle \boldsymbol{p}(t - t'), \mathscr{H}_1(0) \rangle\rangle / E \tag{5}$$

for a crystal dipole moment

$$p = -\sum_n e r_n ,$$

r_n being the displacement of the nth exciton in the lattice. The Green function (5) depends on Green functions of the operators b_k^+ and b_k, and to evaluate these latter functions we will use the equation of motion method (Zubarev 1960). This requires firstly a diagonalization of the Hamiltonian (1), which in matrix form becomes

$$\mathscr{H} = [a_k^+ ; b_k^+] \begin{bmatrix} \mathbf{A} & \mathbf{B} \\ \mathbf{B} & \mathbf{D} \end{bmatrix} \begin{bmatrix} a_k \\ b_k \end{bmatrix}, \tag{6}$$

where

$$[a_k^+ ; b_k^+] = [a_{k_1}^+, ..., a_{k_N}^+ ; b_{k_1}^+, ..., b_{k_N}^+], \qquad \begin{bmatrix} a_k \\ b_k \end{bmatrix} = \begin{bmatrix} a_{k_1} \\ \vdots \\ a_{k_N} \\ b_{k_1} \\ \vdots \\ b_{k_N} \end{bmatrix},$$

$$\mathbf{A} = \begin{bmatrix} \varepsilon_1(k_1)+\gamma N^{-1}, & \gamma N^{-1}, & ..., & \gamma N^{-1} \\ \gamma N^{-1}, & \varepsilon_1(k_2)+\gamma N^{-1}, & ..., & \gamma N^{-1} \\ \vdots & \vdots & & \vdots \\ \gamma N^{-1}, & ... & ..., & \varepsilon_1(k_N)+\gamma N^{-1} \end{bmatrix},$$

$$\mathbf{B} = \begin{bmatrix} g, & 0, & ..., & 0 \\ 0, & g, & ..., & 0 \\ \vdots & \vdots & & \vdots \\ 0, & ... & ..., & g \end{bmatrix}, \qquad \mathbf{D} = \begin{bmatrix} \varepsilon_2, & 0, & ..., & 0 \\ 0, & \varepsilon_2, & ..., & 0 \\ \vdots & \vdots & & \vdots \\ 0, & ... & ..., & \varepsilon_2 \end{bmatrix}.$$

To diagonalize equation (6) we first attempt to diagonalize the submatrix **A**. Note that submatrices **B** and **D** are scalar matrices so that any transformation which will diagonalize **A** will leave **B** and **D** unchanged. The secular equation for **A** is

$$D(\lambda) = \prod_k \{\varepsilon_1(k) - \lambda\} + \gamma N^{-1} \sum_k \prod_{k' \neq k} \{\varepsilon_1(k') - \lambda\} = 0. \tag{7}$$

This is more conveniently written by dividing by the defect-free secular equation

$$D_0(\lambda) = \prod_k \{\varepsilon_1(k) - \lambda\} \tag{8}$$

to give

$$\mathscr{D}(\lambda) = 1 + \gamma N^{-1} \sum_k \{\varepsilon_1(k) - \lambda\}^{-1} = 0. \tag{9}$$

The eigenvectors of **A** have the form

$$S_\lambda = N_\lambda^{-1} \begin{bmatrix} \{\varepsilon_1(k_1) - \lambda\}^{-1} \\ \vdots \\ \{\varepsilon_1(k_i) - \lambda\}^{-1} \\ \vdots \\ \{\varepsilon_1(k_N) - \lambda\}^{-1} \end{bmatrix}, \tag{10}$$

where

$$N_\lambda = \left(\sum_k \{\varepsilon_1(k) - \lambda\}^{-2}\right)^{\frac{1}{2}}. \tag{11}$$

The matrix **S** formed with (10) as columns will then give the transform to diagonalize **A**. Since **A** is symmetric and **S** is normalized, we have that **S** is unitary and so $\mathbf{S}^{-1} = \mathbf{S}^+$, the transpose of **S** (Bellman 1960).

We now form the matrix

$$\mathbf{P} = \begin{bmatrix} \mathbf{S} & \mathbf{0} \\ \mathbf{0} & \mathbf{S} \end{bmatrix}, \tag{12}$$

where **0** is the null matrix. The Hamiltonian (6) may then be written

$$\begin{aligned} \mathscr{H} &= [a_k^+ ; b_k^+]\, \mathbf{P}\mathbf{P}^+ \begin{bmatrix} \mathbf{A} & \mathbf{B} \\ \mathbf{B} & \mathbf{D} \end{bmatrix} \mathbf{P}\mathbf{P}^+ \begin{bmatrix} a_k \\ b_k \end{bmatrix} \\ &= [a_k^+ ; b_k^+]\, \mathbf{P} \begin{bmatrix} \boldsymbol{\lambda} & \mathbf{B} \\ \mathbf{B} & \mathbf{D} \end{bmatrix} \mathbf{P}^+ \begin{bmatrix} a_k \\ b_k \end{bmatrix}, \end{aligned} \tag{13}$$

where $\boldsymbol{\lambda}$ is the diagonal matrix whose diagonal elements are the eigenvalues of **A**, given by equation (9).

The matrix in (13) is now block diagonal and symmetric and may thus be completely diagonalized by treating it as 2×2 matrix. The eigenvalues will then be given by

$$\lambda^{\pm}(\lambda) = \tfrac{1}{2}(\lambda + \varepsilon_2) \pm \tfrac{1}{2}\{(\varepsilon_2 - \lambda)^2 + 4g^2\}^{\frac{1}{2}} \tag{14}$$

(from equation (9), λ has N values) and the transition matrix to diagonalize the matrix in (13), properly normalized, is

$$\mathbf{T} = \begin{bmatrix} \mathbf{T}_{11} & \mathbf{T}_{12} \\ \mathbf{T}_{21} & \mathbf{T}_{22} \end{bmatrix}, \tag{15}$$

where

$$\mathbf{T}_{11} = \begin{bmatrix} \dfrac{x(\lambda_1)+y(\lambda_1)}{[1+\{x(\lambda_1)+y(\lambda_1)\}^2]^{\frac{1}{2}}}, & 0, & \cdot & \cdot & 0 \\ 0, & \cdot & & & \cdot \\ \cdot & & \cdot & & \cdot \\ \cdot & & & \cdot & 0 \\ 0, & \cdot & \cdot & 0, & \dfrac{x(\lambda_N)+y(\lambda_N)}{[1+\{x(\lambda_N)+y(\lambda_N)\}^2]^{\frac{1}{2}}} \end{bmatrix}, \tag{15a}$$

$$\mathbf{T}_{21} = \begin{bmatrix} [1+\{x(\lambda_1)+y(\lambda_1)\}^2]^{-\frac{1}{2}}, & 0, & \cdot & \cdot & 0 \\ 0, & \cdot & & & \cdot \\ \cdot & & \cdot & & \cdot \\ \cdot & & & \cdot & 0 \\ 0, & \cdot & \cdot & 0, & [1+\{x(\lambda_N)+y(\lambda_N)\}^2]^{-\frac{1}{2}} \end{bmatrix}, \tag{15b}$$

$$\mathbf{T}_{12} = \begin{bmatrix} \frac{x(\lambda_1)-y(\lambda_1)}{[1+\{x(\lambda_1)-y(\lambda_1)\}^2]^{\frac{1}{2}}}, & 0, & \cdot & \cdot & 0 \\ 0, & \cdot & & & \cdot \\ \cdot & & \cdot & & \cdot \\ \cdot & & & \cdot & 0 \\ 0, & \cdot & \cdot & 0, & \frac{x(\lambda_N)-y(\lambda_N)}{[1+\{x(\lambda_N)-y(\lambda_N)\}^2]^{\frac{1}{2}}} \end{bmatrix}, \tag{15c}$$

$$\mathbf{T}_{22} = \begin{bmatrix} [1+\{x(\lambda_1)-y(\lambda_1)\}^2]^{-\frac{1}{2}}, & 0, & \cdot & \cdot & 0 \\ 0, & \cdot & & & \cdot \\ \cdot & & \cdot & & \cdot \\ \cdot & & & \cdot & 0 \\ 0, & \cdot & \cdot & 0, & [1+\{x(\lambda_N)-y(\lambda_N)\}^2]^{-\frac{1}{2}} \end{bmatrix}, \tag{15d}$$

with

$$x(\lambda) = (\varepsilon_2 - \lambda)/2g, \qquad y(\lambda) = \{1+x(\lambda)^2\}^{\frac{1}{2}}. \tag{15e}$$

The matrix $\mathbf{T}$ is also unitary, so $\mathbf{T}^{-1} = \mathbf{T}^+$. Thus equation (13) may be written

$$\begin{aligned} \mathscr{H} &= [a_k^+ ; b_k^+]\, \mathbf{P}\mathbf{T}\mathbf{T}^+ \begin{bmatrix} \boldsymbol{\lambda} & \mathbf{B} \\ \mathbf{B} & \mathbf{D} \end{bmatrix} \mathbf{T}\mathbf{T}^+\mathbf{P}^+ \begin{bmatrix} a_k \\ b_k \end{bmatrix} \\ &= [a_k^+ ; b_k^+]\, \mathbf{P}\mathbf{T} \begin{bmatrix} \boldsymbol{\Lambda}^+ & \mathbf{0} \\ \mathbf{0} & \boldsymbol{\Lambda}^- \end{bmatrix} \mathbf{T}^+\mathbf{P}^+ \begin{bmatrix} a_k \\ b_k \end{bmatrix}, \end{aligned} \tag{16}$$

where the $\boldsymbol{\Lambda}^{\pm}$ are diagonal matrices with elements $\lambda^{\pm}(\lambda)$ as given by equation (14).

We can now define new operators $C_{1\lambda}$ and $C_{2\lambda}$ for which the Hamiltonian is diagonal:

$$\begin{bmatrix} C_{1\lambda} \\ C_{2\lambda} \end{bmatrix} = \mathbf{T}^+\mathbf{P}^+ \begin{bmatrix} a_k \\ b_k \end{bmatrix} = \mathbf{R}^+ \begin{bmatrix} a_k \\ b_k \end{bmatrix}, \tag{17}$$

where

$$\mathbf{R} = \mathbf{P}\mathbf{T}. \tag{18}$$

If we expand the square root in equation (14), it will be seen to first order that

$$\lambda^+(\lambda) \approx \varepsilon_2, \qquad \lambda^-(\lambda) \approx \lambda. \tag{19a, b}$$

Hence, since $\mathscr{H}$ in diagonal form is

$$\mathscr{H} = \sum_{\lambda} \{\lambda^+(\lambda)\, C_{1\lambda}^+ C_{1\lambda} + \lambda^-(\lambda)\, C_{2\lambda}^+ C_{2\lambda}\}, \tag{20}$$

we thus generate two bands of energy: one near the exciton energy, which we term the 'exciton band', and one near the magnon energy, the 'magnon band'. It will be seen that the width is influenced by the magnon–exciton interaction, and given by the dispersion formulae (14). Thus one may interpret $C_{1\lambda}$ and $C_{2\lambda}$ as representing

magnon–exciton pairs formed as a result of the interaction g, the operator $C_{1\lambda}$ being exciton-like and $C_{2\lambda}$ being magnon-like.

Putting

$$\mathbf{R} = \begin{bmatrix} \mathbf{R}_{11} & \mathbf{R}_{12} \\ \mathbf{R}_{21} & \mathbf{R}_{22} \end{bmatrix}, \quad \text{then} \quad \mathbf{R}^{-1} = \mathbf{R}^{+} = \begin{bmatrix} \mathbf{R}_{11}^{+} & \mathbf{R}_{21}^{+} \\ \mathbf{R}_{12}^{+} & \mathbf{R}_{22}^{+} \end{bmatrix} \tag{21}$$

and, on performing the multiplication (18), we obtain

$$R_{11}(\boldsymbol{k};\lambda) = \frac{1}{N_\lambda} \frac{x(\lambda)+y(\lambda)}{[1+\{x(\lambda)+y(\lambda)\}^2]^{\frac{1}{2}}} \frac{1}{\varepsilon_1(\boldsymbol{k})-\lambda}, \tag{22a}$$

$$R_{12}(\boldsymbol{k};\lambda) = \frac{1}{N_\lambda} \frac{x(\lambda)-y(\lambda)}{[1+\{x(\lambda)-y(\lambda)\}^2]^{\frac{1}{2}}} \frac{1}{\varepsilon_1(\boldsymbol{k})-\lambda}, \tag{22b}$$

$$R_{21}(\boldsymbol{k};\lambda) = \frac{1}{N_\lambda} \frac{1}{[1+\{x(\lambda)+y(\lambda)\}^2]^{\frac{1}{2}}} \frac{1}{\varepsilon_1(\boldsymbol{k})-\lambda}, \tag{22c}$$

$$R_{22}(\boldsymbol{k};\lambda) = \frac{1}{N_\lambda} \frac{1}{[1+\{x(\lambda)-y(\lambda)\}^2]^{\frac{1}{2}}} \frac{1}{\varepsilon_1(\boldsymbol{k})-\lambda}. \tag{22d}$$

We thus have the complete diagonalization of the Hamiltonian (1), given by the form (20). Equation (5) becomes, in terms of the operators b_k^+ and b_k,

$$G(t-t') = -\alpha^2 N^{-1} \sum_{k,k'} \langle\langle b_k^+(t-t') - b_k(t-t'),\, b_{k'}^+(0) - b_{k'}(0)\rangle\rangle, \tag{23}$$

giving four distinct Green functions. On transforming to the new operators $C_{1\lambda}$ and $C_{2\lambda}$, using equation (17) and its conjugate, we obtain the following Green functions,

$$\begin{aligned}\langle\langle b_k^+(t-t'),\, b_{k'}^+(0)\rangle\rangle &= \sum_{\lambda,\lambda'} \{R_{21}(\boldsymbol{k};\lambda) R_{21}(\boldsymbol{k}';\lambda') \langle\langle C_{1\lambda}^+, C_{1\lambda'}^+\rangle\rangle + R_{21}(\boldsymbol{k};\lambda) R_{22}(\boldsymbol{k}';\lambda') \langle\langle C_{1\lambda}^+, C_{2\lambda'}^+\rangle\rangle \\ &\quad + R_{22}(\boldsymbol{k};\lambda) R_{21}(\boldsymbol{k}';\lambda') \langle\langle C_{2\lambda}^+, C_{1\lambda'}^+\rangle\rangle + R_{22}(\boldsymbol{k};\lambda) R_{22}(\boldsymbol{k}';\lambda') \langle\langle C_{2\lambda}^+, C_{2\lambda'}^+\rangle\rangle\},\end{aligned} \tag{24a}$$

$$\begin{aligned}\langle\langle b_k(t-t'),\, b_{k'}^+(0)\rangle\rangle &= \sum_{\lambda,\lambda'} \{R_{21}(\boldsymbol{k};\lambda) R_{21}(\boldsymbol{k}';\lambda') \langle\langle C_{1\lambda}, C_{1\lambda'}^+\rangle\rangle + R_{21}(\boldsymbol{k};\lambda) R_{22}(\boldsymbol{k}';\lambda') \langle\langle C_{1\lambda}, C_{2\lambda'}^+\rangle\rangle \\ &\quad + R_{22}(\boldsymbol{k};\lambda) R_{21}(\boldsymbol{k}';\lambda') \langle\langle C_{2\lambda}, C_{1\lambda'}^+\rangle\rangle + R_{22}(\boldsymbol{k};\lambda) R_{22}(\boldsymbol{k}';\lambda') \langle\langle C_{2\lambda}, C_{2\lambda'}^+\rangle\rangle\},\end{aligned} \tag{24b}$$

$$\begin{aligned}\langle\langle b_k^+(t-t'),\, b_{k'}(0)\rangle\rangle &= \sum_{\lambda,\lambda'} \{R_{21}(\boldsymbol{k};\lambda) R_{21}(\boldsymbol{k}';\lambda') \langle\langle C_{1\lambda}^+, C_{1\lambda'}\rangle\rangle + R_{21}(\boldsymbol{k};\lambda) R_{22}(\boldsymbol{k}';\lambda') \langle\langle C_{1\lambda}^+, C_{2\lambda'}\rangle\rangle \\ &\quad + R_{22}(\boldsymbol{k};\lambda) R_{21}(\boldsymbol{k}';\lambda') \langle\langle C_{2\lambda}^+, C_{1\lambda'}\rangle\rangle + R_{22}(\boldsymbol{k};\lambda) R_{22}(\boldsymbol{k}';\lambda') \langle\langle C_{2\lambda}^+, C_{2\lambda'}\rangle\rangle\},\end{aligned} \tag{24c}$$

$$\langle\langle b_k(t-t'),\, b_{k'}(0)\rangle\rangle$$

$$= \sum_{\lambda,\lambda'} \{R_{21}(\boldsymbol{k};\lambda)\, R_{21}(\boldsymbol{k}';\lambda')\langle\langle C_{1\lambda}, C_{1\lambda'}\rangle\rangle + R_{21}(\boldsymbol{k};\lambda)\, R_{22}(\boldsymbol{k}';\lambda')\langle\langle C_{1\lambda}, C_{2\lambda'}\rangle\rangle$$

$$+ R_{22}(\boldsymbol{k};\lambda)\, R_{21}(\boldsymbol{k}';\lambda')\langle\langle C_{2\lambda}, C_{1\lambda'}\rangle\rangle + R_{22}(\boldsymbol{k};\lambda)\, R_{22}(\boldsymbol{k}';\lambda')\langle\langle C_{2\lambda}, C_{2\lambda'}\rangle\rangle\}. \tag{24d}$$

The commutation rules for the new operators are obtained from equation (17), e.g.

$$[C_{1\lambda}, C_{1\lambda'}^{+}] = \sum_{k,k'} [R_{11}^{-1}(\boldsymbol{k};\lambda)\, a_k + R_{12}^{-1}(\boldsymbol{k};\lambda)\, b_k,\; R_{11}^{-1}(\boldsymbol{k}';\lambda')\, a_{k'}^{+} + R_{12}^{-1}(\boldsymbol{k}';\lambda')\, b_{k'}^{+}]$$

$$= \sum_{k,k'} \{R_{11}^{-1}(\boldsymbol{k};\lambda)\, R_{11}^{-1}(\boldsymbol{k}';\lambda')[a_k, a_{k'}^{+}] + R_{12}^{-1}(\boldsymbol{k};\lambda)\, R_{12}^{-1}(\boldsymbol{k}';\lambda')[b_k, b_{k'}^{+}]\}$$

$$= \sum_{k} \{R_{11}^{-1}(\boldsymbol{k};\lambda)\, R_{11}^{-1}(\boldsymbol{k};\lambda') + R_{12}^{-1}(\boldsymbol{k};\lambda) R_{12}^{-1}(\boldsymbol{k};\lambda')\} = \delta(\lambda,\lambda'), \tag{25}$$

using the boson commutation rules for a_k and b_k and the fact that they commute with each other. Similarly we have

$$[C_{2\lambda}, C_{2\lambda'}^{+}] = \delta(\lambda,\lambda') \tag{26}$$

and all other commutators are zero.

Using the commutation rules (25) and (26) it is possible to show that all the Green functions on the right-hand sides of equations (24) are zero except $\langle\langle C_{1\lambda}^{+}, C_{1\lambda'}\rangle\rangle$, $\langle\langle C_{1\lambda}, C_{1\lambda'}^{+}\rangle\rangle$, $\langle\langle C_{2\lambda}^{+}, C_{2\lambda'}\rangle\rangle$ and $\langle\langle C_{2\lambda}, C_{2\lambda'}^{+}\rangle\rangle$. The equation of motion of the first of these is

$$i\hbar \frac{\partial}{\partial t}\langle\langle C_{1\lambda}^{+}, C_{1\lambda'}\rangle\rangle = \delta(t)\langle[C_{1\lambda}^{+}(0), C_{1\lambda'}(0)]\rangle - \lambda^{+}(\lambda)\langle\langle C_{1\lambda}^{+}, C_{1\lambda'}\rangle\rangle$$

and, taking a time Fourier transform,

$$\langle\langle C_{1\lambda}^{+}, C_{1\lambda'}\rangle\rangle_{\omega} = -N^{-1}\,\delta(\lambda,\lambda')/\{\hbar\omega + \lambda^{+}(\lambda)\}. \tag{27a}$$

Similarly

$$\langle\langle C_{1\lambda}, C_{1\lambda'}^{+}\rangle\rangle_{\omega} = N^{-1}\,\delta(\lambda,\lambda')/\{\hbar\omega - \lambda^{+}(\lambda)\}, \tag{27b}$$

$$\langle\langle C_{2\lambda}^{+}, C_{2\lambda'}\rangle\rangle_{\omega} = -N^{-1}\,\delta(\lambda,\lambda')/\{\hbar\omega + \lambda^{-}(\lambda)\}, \tag{27c}$$

$$\langle\langle C_{2\lambda}, C_{2\lambda'}^{+}\rangle\rangle_{\omega} = N^{-1}\,\delta(\lambda,\lambda')/\{\hbar\omega - \lambda^{-}(\lambda)\}. \tag{27d}$$

Using the relations (24) and (27), equation (23) becomes

$$G(\omega) = -\alpha^2 N^{-2} \sum_{k,k';\lambda} [R_{21}(\boldsymbol{k};\lambda)\, R_{21}(\boldsymbol{k}';\lambda)\{(\hbar\omega + \lambda^{+}(\lambda))^{-1} - (\hbar\omega - \lambda^{+}(\lambda))^{-1}\}$$

$$+ R_{22}(\boldsymbol{k};\lambda)\, R_{22}(\boldsymbol{k}';\lambda)\{(\hbar\omega + \lambda^{-}(\lambda))^{-1} - (\hbar\omega - \lambda^{-}(\lambda))^{-1}\}]. \tag{28}$$

As the elements of **R** are all real, the optical absorption, which is given by the imaginary part of equation (28), is

$$\chi(\omega) = -\frac{\alpha^2\pi}{N^2}\sum_{k,k';\lambda}[R_{21}(\boldsymbol{k};\lambda)R_{21}(\boldsymbol{k}';\lambda)\{\delta(\hbar\omega+\lambda^+(\lambda))-\delta(\hbar\omega-\lambda^+(\lambda))\}$$
$$+R_{22}(\boldsymbol{k};\lambda)R_{22}(\boldsymbol{k}';\lambda)\{\delta(\hbar\omega+\lambda^-(\lambda))-\delta(\hbar\omega-\lambda^-(\lambda))\}],$$

using

$$\mathrm{Im}\,(x-\mathrm{i}\varepsilon)^{-1}|_{\varepsilon\to 0^+} = \pi\delta(x).$$

As both $\delta(\hbar\omega+\lambda^+(\lambda))$ and $\delta(\hbar\omega-\lambda^+(\lambda))$ cannot be nonzero, we choose the second of these, so that

$$\chi(\omega) = +\frac{\alpha^2\pi}{N^2}\sum_{k,k';\lambda}\{R_{21}(\boldsymbol{k};\lambda)R_{21}(\boldsymbol{k}';\lambda)\delta(\hbar\omega-\lambda^+(\lambda))$$
$$+R_{22}(\boldsymbol{k};\lambda)R_{22}(\boldsymbol{k}';\lambda)\delta(\hbar\omega-\lambda^-(\lambda))\}. \tag{29}$$

The delta function may be written

$$\delta(\hbar\omega-\lambda^\pm(\lambda)) = \delta(\lambda-\lambda_\nu)/|\lambda'^\pm(\lambda)|, \tag{30}$$

where the prime denotes differentiation with respect to λ here, and λ_ν satisfies the relation $\hbar\omega-\lambda^\pm(\lambda_\nu) = 0$, or

$$\lambda_\nu = \{\hbar\omega(\hbar\omega-\varepsilon_2)-g^2\}/(\hbar\omega-\varepsilon_2) \tag{31}$$

using equation (14).

We can sum (29) over λ if we use the density of states function $g(\lambda)$ defined by (Callaway 1964)

$$g(\lambda) = \pi^{-1}\,\mathrm{Im}\frac{\mathrm{d}}{\mathrm{d}\lambda}\Big(\ln D(\lambda)\Big) = \pi^{-1}\,\mathrm{Im}\frac{\mathrm{d}}{\mathrm{d}\lambda}\Big(\ln\mathscr{D}(\lambda)+\ln D_0(\lambda)\Big), \tag{32}$$

from equations (7) and (9). After summation over λ, equation (29) then becomes

$$\chi(\omega) = \frac{\alpha^2\pi}{N^2}\sum_{k,k'}\{R_{21}(\boldsymbol{k};\lambda_\nu)R_{21}(\boldsymbol{k}';\lambda_\nu)g(\lambda_\nu)|\lambda'^+(\lambda_\nu)|^{-1}$$
$$+R_{22}(\boldsymbol{k};\lambda_\nu)R_{22}(\boldsymbol{k}';\lambda_\nu)g(\lambda_\nu)|\lambda'^-(\lambda_\nu)|^{-1}\}. \tag{33}$$

However, from equation (14)

$$\lambda'^\pm(\lambda) = \tfrac{1}{2}\left(1\pm\frac{-(\varepsilon_2-\lambda)}{\{(\varepsilon_2-\lambda)^2+4g^2\}^{\frac{1}{2}}}\right) \tag{34}$$

and using (31) it can be shown that, in equations (22c) and (22d) respectively,

$$\lambda'^+(\lambda_\nu) = 1+\{x(\lambda)+y(\lambda)\}^2 \quad \text{and} \quad \lambda'^-(\lambda_\nu) = 1+\{x(\lambda)-y(\lambda)\}^2,$$

where $x(\lambda)$ and $y(\lambda)$ are as defined by equations (15e). Thus (33) becomes

$$\chi(\omega) = \frac{\alpha^2\pi}{N^2}\sum_{k,k'}\left(\frac{1}{N_{\lambda_\nu}^2}\frac{1}{\varepsilon_1(\boldsymbol{k})-\lambda_\nu}\frac{1}{\varepsilon_1(\boldsymbol{k}')-\lambda_\nu}g(\lambda_\nu)\right). \tag{35}$$

The ranges for the magnon and exciton bands are given in equation (35) by the ranges for which $g(\lambda_v)$ is nonzero. The 'magnon sideband' is considered to be the exciton band in this expression. Apart from these two bands, equation (35) allows the possibility of local modes produced by the presence of the defect. These will be represented as δ-functions at frequencies outside the bands given by

$$|D(\lambda)| = 0$$

(Maradudin *et al.* 1963). When the number of defects is n, this may be accounted for by summing over all defect sites. If we assume that all defects are identical, we may introduce the concentration $C = n/N$ of defects into the first term of equation (32), provided that C is sufficiently small for the interaction between the defects to be ignored.

For the ideal crystal, with $\gamma = 0$, the matrix **P** becomes the identity matrix, as **A** in equation (6) is already diagonal, so that the sum of any column of the submatrix **S** of **P** is 1, that is,

$$N_\lambda^{-1} \sum_{\boldsymbol{k}} \{\varepsilon_1(\boldsymbol{k}) - \lambda\}^{-1} \to 1 \qquad \text{as} \qquad \gamma \to 0. \tag{36}$$

Hence in this case

$$\chi^0(\omega) = (\alpha^2 \pi/N) g_0(\lambda_v), \tag{37}$$

where $g_0(\lambda)$ is the density of states function defined from the secular determinant (8).

It should be pointed out that although we have used the external electric field magnitude E in expressions for $\chi(\omega)$, this will not be the actual field present at the exciton dipole. The true field is that which is modified by the environment of the exciton and would more accurately be given by

$$E' = \boldsymbol{\varepsilon} . \boldsymbol{E},$$

for some dielectric tensor $\boldsymbol{\varepsilon}$.

One-dimensional Model

To illustrate the properties of the results obtained above, we consider a one-dimensional crystal, or equivalently a crystal in which the magnons propagate in one dimension only. For this problem we can obtain an exact analytic solution.

Now there are two nearest neighbours, $z = 2$, and equation (2) becomes

$$\varepsilon_1(\boldsymbol{k}) = \varepsilon_0(1 - \alpha \cos k), \tag{38}$$

where

$$\varepsilon_0 = 2JSz(1 - S\rho), \qquad \alpha = (1 - S\rho)^{-1}. \tag{39}$$

The secular determinant (9) may now be evaluated by changing the sum to an integral to give

$$\mathscr{D}(\lambda) = 1 + i\gamma / \{\alpha^2 \varepsilon_0^2 - (\varepsilon_0 - \lambda)^2\}^{\frac{1}{2}}, \tag{40}$$

for

$$\alpha^2 \varepsilon_0^2 > (\varepsilon_0 - \lambda)^2. \tag{41}$$

Thus using equation (32)

$$g(\lambda) = \frac{\gamma n |\varepsilon_0 - \lambda|}{\pi \{\alpha^2 \varepsilon_0^2 - (\varepsilon_0 - \lambda)^2\}^{\frac{1}{2}} \{\alpha^2 \varepsilon_0^2 + \gamma^2 - (\varepsilon_0 - \lambda)^2\}} + g_0(\lambda), \tag{42}$$

with

$$g_0(\lambda) = N/\pi\{\alpha^2\varepsilon_0^2-(\varepsilon_0-\lambda)^2\}^{\frac{1}{2}}, \tag{43}$$

and $g(\lambda)$ is only nonzero for the condition (41), apart from any local modes. Use of equation (31) in (41) gives the ranges of the two bands, for $\alpha > 0$:

$$\frac{\varepsilon_0(1-\alpha)-\varepsilon_2}{2}\left\{1-\left(1+\frac{g^2}{\frac{1}{4}\{\varepsilon_0(1-\alpha)-\varepsilon_2\}^2}\right)^{\frac{1}{2}}\right\}$$

$$< \hbar\omega-\varepsilon_2 < \frac{\varepsilon_0(1+\alpha)-\varepsilon_2}{2}\left\{1-\left(1+\frac{g^2}{\frac{1}{4}\{\varepsilon_0(1+\alpha)-\varepsilon_2\}^2}\right)^{\frac{1}{2}}\right\} \tag{44}$$

and

$$\frac{\varepsilon_0(1-\alpha)-\varepsilon_2}{2}\left\{1+\left(1+\frac{g^2}{\frac{1}{4}\{\varepsilon_0(1-\alpha)-\varepsilon_2\}^2}\right)^{\frac{1}{2}}\right\}$$

$$< \hbar\omega-\varepsilon_2 < \frac{\varepsilon_0(1+\alpha)-\varepsilon_2}{2}\left\{1+\left(1+\frac{g^2}{\frac{1}{4}\{\varepsilon_0(1+\alpha)-\varepsilon_2\}^2}\right)^{\frac{1}{2}}\right\}. \tag{45}$$

For $\alpha < 0$ the inequalities are reversed. Then, (44) gives the range of the exciton band and (45) the range of the magnon band. It is the former which is usually termed the magnon sideband.

From equations (9) and (11) we have

$$N_\lambda^2 = \sum_{\boldsymbol{k}}\left(\frac{1}{\varepsilon_1(\boldsymbol{k})-\lambda}\right)^2 = \frac{N}{\gamma}\frac{\mathrm{d}|\mathscr{D}(\lambda)|}{\mathrm{d}\lambda},$$

and so, using equation (40),

$$N_\lambda^2 = N|\varepsilon_0-\lambda|/\{\alpha^2\varepsilon_0^2-(\varepsilon_0-\lambda)^2\}^{3/2}. \tag{46}$$

Then equation (35) becomes, for a one-dimensional model,

$$\chi(\omega) = \frac{\alpha^2\pi}{N\gamma^2}\frac{\{\alpha^2\varepsilon_0^2-(\varepsilon_0-\lambda_\nu)^2\}^{3/2}}{|\varepsilon_0-\lambda_\nu|}g(\lambda_\nu) \tag{47}$$

for the two ranges (44) and (45).

The presence of the defect will also contribute some local modes which lie outside the ranges of (44) and (45). Such modes will be represented as delta functions, of strength equal to the concentration C of the defect, and at frequencies ω given by the solutions of

$$1-\gamma/\{(\varepsilon_0-\lambda_\nu)^2-\alpha^2\varepsilon_0^2\}^{\frac{1}{2}} = 0,$$

which are, from equation (31)

$$\hbar\omega-\varepsilon_2 = \frac{\varepsilon_0+(\alpha^2\varepsilon_0^2+\gamma^2)^{\frac{1}{2}}-\varepsilon_2}{2}\left\{1\pm\left(1+\frac{g^2}{\frac{1}{4}\{\varepsilon_0+(\alpha^2\varepsilon_0+\gamma^2)^{\frac{1}{2}}-\varepsilon_2\}^2}\right)^{\frac{1}{2}}\right\}. \tag{48}$$

It will be seen that the frequencies given by (48) lie outside the two bands (44) and (45), and that one local mode is just above the magnon band and one just above the exciton band. The separation from the band is dependent on γ, the defect energy.

A plot of the two line profiles is shown in Fig. 1 for the specified values of the parameters $\varepsilon_0/\varepsilon_2$, γ/ε_2, g^2/ε_2^2, α and concentration C of the defect. The frequency scale is given in terms of ε_2. It is seen from the figure that the effect of introducing an imperfection into the lattice is to dramatically alter the excitonic spectrum from that of a perfect lattice by shifting the square root discontinuity from the edges of the two branches of the spectrum to approximately the centre of each branch (the case when $\gamma \to 0$ is discussed above). The displacement of the discontinuities from

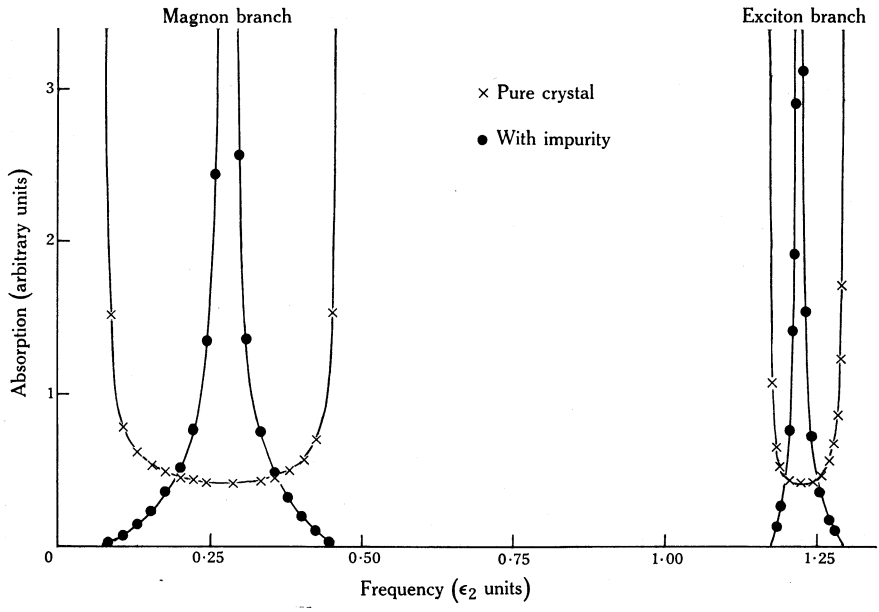

Fig. 1. Excitonic spectrum of a one-dimensional ferromagnet with impurity, of concentration $C = 0{\cdot}5$, for the parameters $\varepsilon_0/\varepsilon_2 = 0{\cdot}5$, $\gamma/\varepsilon_2 = 0{\cdot}01$, $g^2/\varepsilon_2^2 = 0{\cdot}16$ and $\alpha = 0{\cdot}5$. The exciton branch contains the 'magnon sideband'. The frequencies are given in units of the exciton energy ε_2. For the parameters chosen, the branches have the ranges: magnon, $0{\cdot}076707\,\varepsilon_2$ to $0{\cdot}455924\,\varepsilon_2$; exciton, $1{\cdot}173293\,\varepsilon_2$ to $1{\cdot}294076\,\varepsilon_2$.

the exact centre of each branch is caused by the interaction g, the magnon peak being shifted to higher frequencies and the exciton peak to lower frequencies. The effect of g may be seen from Fig. 2, which shows the dependence of the exciton branch line profile on g^2/ε_2^2 (in this figure the branch has been normalized to the frequency range 0 to 1). It is this branch which is studied in optical absorption experiments and which is known as the 'magnon sideband'. The magnon branch frequency is normally of the order of $10\,\mathrm{cm}^{-1}$ and is thus outside the scope of most optical absorption apparatus, while the exciton branch frequency is in the infrared or optical region and more readily studied.

Fig. 2 shows that, as the interaction g is reduced, the exciton branch becomes more skewed. At the same time, the result (44) shows that the width of the branch reduces to zero, so that at $g = 0$ we have only a delta function at the exciton frequency. The magnon branch becomes symmetric in the range 0 to 1 as g^2/ε_2^2 approaches zero.

It should be noted that the spectrum of Fig. 1 should include two local modes at frequencies given by equation (48). There will be one at a frequency just above the

magnon branch (here at $0{\cdot}45605\,\varepsilon_2$) and one just above the exciton branch ($1{\cdot}29414\,\varepsilon_2$). As pointed out earlier, the separation of the local mode from the branch depends on the defect energy γ, approaching the upper limit of the branch as γ approaches zero.

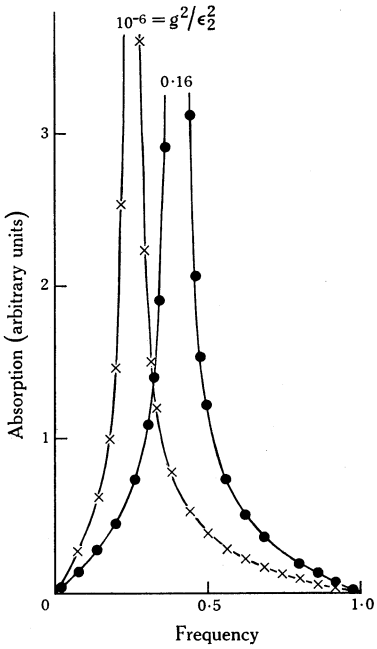

Fig. 2. Dependence of the exciton branch spectrum on the interaction g, as illustrated by the variation in the profile for a change in the parameter g^2/ε_2^2 from $0{\cdot}16$ to 10^{-6}. As g^2/ε_2^2 decreases the curve becomes more skewed, while the width of the branch reduces to zero (from equation (44)). The other parameters here are as in Fig. 1, except that the impurity concentration C is $0{\cdot}01$. Note that the frequency range is normalized to (0, 1).

Three-dimensional Model

A comprehensive quantitative account of the calculation of the excitonic spectrum of a three-dimensional crystal will be given in a future publication, but a brief qualitative discussion is included here. We consider a simple cubic crystal with the vector Δ representing the positions of nearest neighbours in the crystal, and $|\Delta_1| = |\Delta_2| = |\Delta_3| = a$ the crystal lattice constant. Equation (2) now becomes

$$\varepsilon_1(\boldsymbol{k}) = 2JSz\{1-2z^{-1}(\cos k_1 a + \cos k_2 a + \cos k_3 a)\} - 2zJS^2\rho\,, \tag{49}$$

for $\boldsymbol{k} = (k_1, k_2, k_3)$ and $z = 6$.

The difficulty with three-dimensional calculations arises from the evaluation of the sum in equation (9), which leads to a lattice Green function (for a simple cubic crystal) of the form (Mahanty 1966)

$$G(\boldsymbol{j}_1, \boldsymbol{j}_2; z) = \frac{1}{V^*}\int_{V^*} \frac{\mathrm{d}^3k\, \exp\{\mathrm{i}\boldsymbol{k}.(\boldsymbol{j}_1-\boldsymbol{j}_2)\}}{\varepsilon(\boldsymbol{k})-z}, \tag{50}$$

where the energy band dispersion $\varepsilon(\boldsymbol{k})$ is given by equation (49) and V^* is the volume of the first Brillouin zone. There are several ways of evaluating equation (50). Joyce (1973) has given expansions in terms of hypergeometric functions while Mahanty (1966) has described a Fourier series expansion. For the present case it is more convenient to follow Mahanty's work.

Using the Dirac relation

$$\lim_{\varepsilon \to 0^+} (x \pm \mathrm{i}\varepsilon)^{-1} = P/x \mp \mathrm{i}\pi\,\delta(x),$$

equation (9) may be written (with N replaced by N^3 for a three-dimensional crystal)

$$\mathscr{D}(\lambda) = 1 + \gamma R(\lambda) + \mathrm{i}\pi\gamma g_0(\lambda), \tag{51}$$

where

$$R(\lambda) = \frac{P}{N^3} \int \frac{\mathrm{d}\boldsymbol{k}}{\varepsilon_1(\boldsymbol{k}) - \lambda} \tag{52}$$

and

$$g_0(\lambda) = \delta(\varepsilon_1(\boldsymbol{k}) - \lambda). \tag{53}$$

Equation (53) is the pure-crystal density of states. The quantities $R(\lambda)$ and $g_0(\lambda)$ are the real and imaginary parts of the lattice Green function (50), and Mahanty (1966) has given Fourier series expansions for them, where $g_0(\lambda) = \operatorname{Im} G(\boldsymbol{0}; x)$ and $R(\lambda) = \operatorname{Re} G(\boldsymbol{0}; x)$.

From equations (32) and (51), we have

$$\begin{aligned} g(\lambda) &= \pi^{-1} \operatorname{Im} \left| \frac{\gamma R'(\lambda) + \mathrm{i}\pi\gamma g_0'(\lambda)}{1 + \gamma R(\lambda) + \mathrm{i}\pi\gamma g_0(\lambda)} \right| + g_0(\lambda) \\ &= \gamma \left| \frac{g_0'(\lambda)\{1 + \gamma R(\lambda)\} - \gamma R'(\lambda) g_0(\lambda)}{\{1 + \gamma R(\lambda)\}^2 + \{\pi\gamma g_0(\lambda)\}^2} \right| + g_0(\lambda), \end{aligned} \tag{54}$$

where the prime denotes differentiation with respect to λ. As for the one-dimensional case, the coefficient of $g(\lambda_\nu)$ in equation (35) may be expressed in terms of $\mathscr{D}(\lambda)$ so that

$$\frac{1}{N_\lambda^2} \sum_{\boldsymbol{k},\boldsymbol{k}'} \frac{1}{\varepsilon_1(\boldsymbol{k}) - \lambda} \frac{1}{\varepsilon_1(\boldsymbol{k}') - \lambda} = \frac{N^3}{\gamma^2} \frac{1}{|R'(\lambda) + \mathrm{i}\pi g_0'(\lambda)|}. \tag{55}$$

Substitution of (54) and (55) into (35) then gives the solution of the problem (with N replaced by N^3).

All the functions in equations (54) and (55) may be evaluated by Fourier series. It can be shown, however, that $g_0(\lambda)$ and $R(\lambda)$ have cusp points (the infinite discontinuities disappear), so that at these points $g_0'(\lambda)$ and $R'(\lambda)$ are not defined. Nevertheless, it is expected that these latter functions will be well behaved in the intervals between the cusp points, thus enabling a numerical determination of $\chi(\omega)$.

The condition that $g(\lambda)$ be nonzero again yields two branches of the excitonic spectrum, and in addition there will be local modes at frequencies determined by

$$|\mathscr{D}(\lambda)| = 0, \tag{56}$$

outside the magnon and exciton branches. The existence of solutions and their values will again depend on γ.

Acknowledgment

The author would like to express gratitude to Dr J. Mahanty for many useful suggestions during the course of this work.

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Manuscript received 7 March 1974