

Spin-Wave Impurity States in Ferromagnets

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The effect of dilute impurities on the spin-wave spectrum of ferromagnetic insulators has been studied. The theory of spin-wave impurity states is developed from the Heisenberg exchange Hamiltonian using Green's function techniques. The low-lying, *s*-like, impurity states for cubic crystals are discussed and shown to depend only upon the unperturbed spin-wave density of states. Numerical results are obtained for simple cubic crystals. It is shown that localized states lying outside of the spin-wave band as well as virtual states which decay into the continuum can exist. The energy of the *s*-, *p*-, and *d*-like states is obtained as a function of the ratios of the impurity spin and effective exchange to those of the host atoms. The nature of the virtual states is discussed and a general expression for the width is obtained by considering the change in the density of states due to the impurities. Low-lying, *s*-like, virtual states with long lifetimes are found to exist, and it is suggested that these states may cause significant effects in the spin-wave specific heat and thermal conductivity of impure ferroinsulators.

I. INTRODUCTION

THE effect of impurities on the spectrum of elementary excitations of a solid has received a great deal of attention in recent years. This subject is particularly interesting to the theorist because within the quasiparticle approximation the effect of a localized perturbation in a periodic structure can be worked out exactly using Green's function techniques. The perturbation may be very large in this class of problems: The condition for solubility is that the impurity perturbation be short range. The principal result of the impurity problem is that the perturbed region surrounding the impurity appears as a separate system whose energy levels depend upon the energy spectrum of the host crystal. If the perturbation is sufficiently large, an impurity energy level may lie outside of the energy band or continuum of the host. In this case, the impurity state will have a very long lifetime since there are no host states in the immediate energy range to facilitate decay. These are the bound or localized impurity states. On the other hand, if the energy lies in the host energy band then the impurity state is virtual since it will decay into the continuum with a finite lifetime. These virtual states may, however, be important since the lifetime may be long compared to the characteristic relaxation time of some physical processes. In general, bound or virtual states can result in any quasiparticle system because of the presence of impurities.

The effect of an impurity atom on the lattice vibrations of a solid has been discussed by Montroll and Potts¹ and more recently by Mahanty, Maradudin, and Weiss.² The chief result of these investigations is that localized vibrational modes outside of the continuum of the phonon energy band can result if the impurity

mass is lighter than the host mass. Virtual states which decay into the continuum can result for impurity mass greater than that of the host. It has been recently pointed out that it may be possible to observe virtual states using the Mössbauer effect.^{3,4}

Most of the work concerning the localized mode has treated in detail only the case of a change in mass—the “isotopic impurity problem.” Visscher,⁵ however, has considered the effect of changes in force constants as well as mass for a simplified nearest neighbor lattice dynamics model.

In the electronic problem the counterpart of the localized mode is the bound electron impurity state. Koster and Slater⁶ have treated this problem in some detail. They show that if the electronic perturbation due to the impurity is sufficiently large electronic states with wave functions localized about the impurity can result with energies above (or below) the electron energy band. The effect of states in the continuum is, however, discussed only from a scattering point of view. Electronic virtual states have also been discussed⁷⁻⁹ in connection with the occurrence of localized magnetic states in metals.

In magnetic solids at low temperatures one must consider in addition to the electron and phonon excitations, the excitation of spin waves.¹⁰ Spin waves are, of course, a part of the electron excitation spectrum. In the ideal insulating magnetic solid the magnon energy

³ R. Brout and W. Visscher, *Phys. Rev. Letters* **9**, 54 (1962).

⁴ G. W. Lehman and R. E. DeWames, *Phys. Rev. Letters* **9**, 344 (1962).

⁵ W. Visscher, *Phys. Rev.* **129**, 28 (1963).

⁶ G. F. Koster and J. C. Slater, *Phys. Rev.* **96**, 1208 (1954).

⁷ P. A. Wolff, *Phys. Rev.* **124**, 1030 (1961); and A. M. Clogston, *ibid.* **125**, 439 (1962).

⁸ P. W. Anderson, *Phys. Rev.* **124**, 41 (1961).

⁹ A. Blandin and J. Friedel, *J. Phys. Radium* **19**, 573 (1958).

¹⁰ For a review of spin wave theory see, for example, A. I. Akhiezer, V. G. Bar'yakhtar, and M. I. Kagnov, *Usp. Fiz. Nauk* **71**, 533 (1960) [translation: *Soviet Phys.—Usp.* **3**, 567 (1961)].

¹ E. W. Montroll and R. B. Potts, *Phys. Rev.* **100**, 525 (1955).

² J. Mahanty, A. A. Maradudin, and G. H. Weiss, *Progr. Theoret. Phys. (Kyoto)* **20**, 369 (1958).

band will be separated from the excited one-electron states by an energy gap¹¹ so that it is meaningful to treat the magnetization fluctuations as a separate system of elementary excitations. At sufficiently low temperatures magnons obey Bose statistics and behave like phonons. By analogy one might expect that localized spin-wave modes due to impurities could result. This, in fact, is the case. It is worth pointing out, however, that there is no spin wave analog of the phonon 'isotopic impurity problem.' This is because a change in the spin of the impurity changes the torques exerted on all the neighboring spins to which it is coupled by exchange. Thus, the impurity problem for the magnetic solid is like the lattice vibration problem in which both the mass and the effective atomic force constants are changed. The analogy, however, is not complete and the problems differ in several important respects. Since exchange is an electronic interaction, we also expect that the formal structure of the theory of electron impurity states should apply to the spin-wave impurity problem. This conjecture turns out to be true and much of our analysis is similar to that employed by Koster and Slater.⁶ However, there are several special features peculiar to the spin-wave impurity problem which do not obtain in the electronic problem.

In this paper, we consider the effect of a single substituted impurity atom on the spin-wave spectrum of an insulating ferromagnet. The impurity atom's spin and effective exchange will, in general, be different from those of the host atoms. We assume that the magnetic properties of the system at low temperatures are described by the simple Heisenberg spin Hamiltonian with nearest neighbor exchange interactions only. It is worth mentioning in connection with this assumption that there does exist a class of ferromagnetic insulators to which the Heisenberg Hamiltonian applies phenomenologically. Several rare-earth binary compounds, of which EuS is a typical example, have been investigated recently¹²⁻¹⁴ and found to be ferromagnetic at low temperatures (on the order of 5-80°K). Magnon-magnon interactions are at a minimum in these ferroinsulators because of the large atomic spins associated with the rare earths. In addition, the rare-earth ions are frequently found in an *s* state so that the complications due to orbital angular momentum are absent. EuS, for example, has a Curie temperature about 18°K and the europium ion is in an *s* state with spin $\frac{7}{2}$. The spin-wave contribution to the specific heat has been examined¹² by McCollum and Callaway between 1 and 4°K. The agreement between the Heisenberg theory of spin waves and the experimental results is impressive.

¹¹ C. Herring and C. Kittel, Phys. Rev. **81**, 869 (1951).

¹² T. R. McGuire, B. E. Argyle, M. W. Shafer, and J. S. Smart, Appl. Phys. Letters **1**, 17 (1962).

¹³ D. C. McCollum, Jr., and J. Callaway, Phys. Rev. Letters **9**, 376 (1962).

¹⁴ B. T. Matthias, R. M. Bozorth, and J. H. Van Vleck, Phys. Rev. Letters **7**, 160 (1960).

In Sec. II, the general theory of spin-wave impurity states is developed. The Heisenberg Hamiltonian for spin waves in an unperturbed ferroinsulator is transformed into the Schrödinger representation and diagonalized. Only nearest neighbor interactions are included. The Green's function for spin waves is derived. The Schrödinger equation for a ferromagnet with a single substituted impurity atom is developed. The impurity problem is then solved in terms of the matrix elements of the Green's function. In Sec. III, application of the theory to cubic crystals is considered. The nature of the low-lying, *s*-like, impurity states is investigated. A formula is derived relating the impurity state energy to the changes in spin and exchange. It is shown that the *s*-like states are determined entirely by the density of states. The analysis of all the impurity states (*s*, *p*, and *d*) for a simple cubic crystal is then given. The energy of the impurity state is shown as a function of the change in the spin and exchange. Section IV is concerned with the general features of resonant or virtual impurity states. A general expression for the width of a virtual state is developed. In Sec. V, the locations and widths of the spin-wave impurity states for the simple cubic crystal are considered in detail.

II. THE THEORY OF SPIN-WAVE IMPURITY STATES

1. Schrödinger Equation for Spin Waves

The calculation of the energies of spin-wave modes associated with impurities is easiest if we use an effective Schrödinger equation for the spin waves. This representation is analogous to the use of Wannier functions in energy band theory. In this section the Schrödinger equation for unperturbed spin waves in a ferromagnetic insulator is derived. The simple isotropic Heisenberg exchange Hamiltonian is

$$\mathcal{H} = -J \sum_j \sum_{\Delta} \mathbf{S}(j) \cdot \mathbf{S}(j+\Delta) - H_z g \beta \sum_j \mathbf{S}(j)_z, \quad (1)$$

where J is the effective nearest neighbor exchange, H_z is an external magnetic field assumed to be along the z direction, g is the spectroscopic splitting factor, and β is the Bohr magneton. $\mathbf{S}(j)$ is the atomic spin operator for the atom located at the lattice position $\mathbf{R}(j)$ and $\mathbf{S}(j)_z$ is its z component. The sum over Δ is a sum over the z vectors which connect an atom with its z nearest neighbors. The first term in Eq. (1) describes the energy due to inter-atomic exchange and the second term is the Zeeman energy for the system of spins in an external magnetic field. The maximum value of $\mathbf{S}(j)_z$ is S , the spin quantum number.

It is convenient to introduce the spin annihilation and creation operators \mathbf{S}^- and \mathbf{S}^+

$$\mathbf{S}(j)_{\pm} = \mathbf{S}(j)_{x \pm i y} \quad (2)$$

which obey the commutator relations

$$\begin{aligned} [\mathbf{S}(j)^+, \mathbf{S}(k)^-] &= 2\delta_{jk}\mathbf{S}(j)_z, \\ [\mathbf{S}(j)_z, \mathbf{S}(k)^-] &= -\delta_{jk}\mathbf{S}(j)^-, \\ [\mathbf{S}(j)_z, \mathbf{S}(k)^+] &= \delta_{jk}\mathbf{S}(j)^+. \end{aligned} \quad (3)$$

Equation (1) may be expressed in terms of these operators with the result

$$\begin{aligned} &= -J \sum_j \sum_{\Delta} \{\mathbf{S}(j)_z \mathbf{S}(j+\Delta)_z + \mathbf{S}(j+\Delta)^- \mathbf{S}(j)^+\} \\ &\quad - H_z g \beta \sum_j \mathbf{S}(j)_z. \end{aligned} \quad (4)$$

In this paper, we neglect magnon-magnon interactions and, therefore, need only consider the single quasi-particle states. The ground state or vacuum of our ferromagnet corresponding to the state of complete spin alignment is denoted by $|0\rangle$ and defined by the condition that

$$\mathbf{S}(j)^+ |0\rangle = 0, \quad (5)$$

for all j . We may then define a complete set of orthonormal single spin deviation states^{15,16}

$$|j\rangle = (2S)^{-1/2} \mathbf{S}(j)^- |0\rangle, \quad (6)$$

where S is the spin quantum number. The operator $\mathbf{S}(j)^-$ operating on the vacuum lowers the z component of the atomic spin at the j th site by one unit and, therefore, 'creates' a spin deviation. These states are analogous to the Wannier functions of band theory. The kets, $|j\rangle$, are the basis vectors of an N -dimensional Hilbert space. An arbitrary one-particle state may be expanded in terms of these kets,

$$\Psi = \sum_j \varphi(j) |j\rangle, \quad (7)$$

where the $\varphi(j)$ is the projection of Ψ on the basis vector $|j\rangle$ and $|\varphi(j)|^2$ may be interpreted as the probability of a spin deviation on the j th atomic site. Thus, a Schrödinger wave function is an N -row column,

$$\mathbf{U} = \begin{pmatrix} \varphi(1) \\ \varphi(2) \\ \vdots \\ \varphi(N) \end{pmatrix}. \quad (8)$$

The operator equation,

$$\mathcal{H}\Psi = E\Psi, \quad (9)$$

can be transformed into a Schrödinger equation for the amplitudes φ by operating from the left with the bra, $\langle k|$, with the result that

$$\sum_j \langle k | \mathcal{H} | j \rangle \varphi(j) = E \varphi(k). \quad (10)$$

Equation (10) is the Schrödinger equation for spin waves. The matrix elements are easily obtained using Eqs. (4), (7), and (3):

$$\langle k | \mathcal{H} | j \rangle = (E^0 + H_z g \beta + 2JSZ) \delta_{jk} - 2JS \sum_{\Delta} \delta_{j+\Delta, k}, \quad (11)$$

where

$$E^0 = -JNS^2Z - Ng\beta H_z S. \quad (12)$$

Equation (10) can now be written as

$$E \varphi(k) = (E^0 + H_z g \beta + 2JSZ) \varphi(k) - 2JS \sum_{\Delta} \varphi(k+\Delta). \quad (13)$$

Introducing the N -dimensional column vector \mathbf{U} we have

$$E\mathbf{U} = \mathbf{H}^0 \mathbf{U}, \quad (14)$$

where \mathbf{H}^0 is an $N \times N$ matrix with matrix elements given by Eq. (11). Equation (14) may be recognized as the matrix equation for a lattice of coupled oscillators. The eigenvectors are, therefore,

$$\mathbf{U}_q = N^{-1/2} \begin{pmatrix} \exp\{i\mathbf{q} \cdot \mathbf{R}(1)\} \\ \exp\{i\mathbf{q} \cdot \mathbf{R}(2)\} \\ \vdots \\ \exp\{i\mathbf{q} \cdot \mathbf{R}(N)\} \end{pmatrix}, \quad (15)$$

with eigenvalue

$$E_q = E^0 + H_z g \beta + 2JS \left[z - \sum_{\Delta} \cos\{\mathbf{q} \cdot \mathbf{R}(\Delta)\} \right]. \quad (16)$$

The wave function \mathbf{U}_q is a spin wave and E_q is the quantum of energy or magnon associated with it. The spin wave propagation vectors \mathbf{q} are determined in the usual way by the periodic boundary conditions appropriate to the crystal structure. Since the magnetic field shifts all the energies by the same amount it may for the purposes of our discussion be omitted.

2. Green's Function for Spin Waves

In this section, we wish to determine the matrix elements of the operator $1/(E - \mathbf{H}^0)$, which is the Green's function, in the representation defined by the kets $|j\rangle$. The elements of $(E - \mathbf{H}^0)^{-1}$ are $(E - E_q)^{-1} \delta_{q,q'}$ in the system in which \mathbf{H}^0 is diagonal. The transformation between these systems is accomplished by means of the unitary transformation, D , constructed from the columns \mathbf{U}_q ,

$$D = (\mathbf{U}_{q_1} \mathbf{U}_{q_2} \cdots \mathbf{U}_{q_N}). \quad (17)$$

The matrix elements of $(E - \mathbf{H}^0)^{-1}$ are easily obtained

$$\begin{aligned} &\{(E - \mathbf{H}^0)^{-1}\}_{ij} \\ &= \sum_q \sum_{q'} D_{iq} \{(E - \mathbf{H}^0)^{-1}\}_{q'q} D_{q'j}^\dagger \\ &= N^{-1} \sum_q \sum_{q'} \exp\{i\mathbf{q} \cdot \mathbf{R}(i) - i\mathbf{q}' \cdot \mathbf{R}(j)\} \delta_{q,q'} (E - E_q)^{-1} \\ &= N^{-1} \sum_q \exp[i\mathbf{q} \cdot \{\mathbf{R}(i) - \mathbf{R}(j)\}] (E - E_q)^{-1}, \end{aligned} \quad (18)$$

where s denotes spectral representation.

¹⁵ F. Bloch, Z. Physik **61**, 206 (1930); **74**, 295 (1932).

¹⁶ The effect of magnon-magnon interactions has been considered by Dyson. F. J. Dyson, Phys. Rev. **102**, 1217 (1956).

It is convenient to work with the dimensionless Green's function \mathbf{G} which is defined by

$$G_{ij} = -4JS[(E - \mathbf{H}^0)^{-1}]_{ij}. \quad (19)$$

Defining

$$\begin{aligned} \mathcal{E} &= (E - E^0 + 2JSZ)(-4JS)^{-1}, \\ \mathcal{E}_q &= \frac{1}{2} \sum_{\Delta} \cos\{\mathbf{q} \cdot \mathbf{R}(\Delta)\}, \end{aligned} \quad (20)$$

then

$$G_{ij} = N^{-1} \sum_q \exp[i\mathbf{q} \cdot \{\mathbf{R}(i) - \mathbf{R}(j)\}] (\mathcal{E} - \mathcal{E}_q)^{-1}. \quad (21)$$

3. Schrödinger Equation for the Perturbed Ferromagnet

In this section we develop the Schrödinger equation for a ferromagnet with a single substituted impurity. The techniques used in Sec. 2 can be applied. Let S' be the spin of the impurity atom and J' the effective exchange between the impurity and a nearest neighbor host atom. The vacuum state, $|0\rangle$ for this system is the state in which the host and impurity spins are all aligned. We can define a complete set of one-particle states for this system similar to those defined for the unperturbed ferromagnet:

$$|j\rangle = \{2S(j)\}^{-1/2} \mathbf{S}(j)^{-} |0\rangle, \quad (22)$$

where $S(j) = S$ for a host atom or S' for the impurity. These states are orthogonal to each other and to the vacuum and properly normalized. We emphasize again that $|0\rangle$ is the true vacuum for the impure ferromagnet. The impurity spin operator obeys the same commutation relation as the host spin operators and, therefore, we may proceed just as in Sec. 2 and obtain the Schrödinger equation

$$\begin{aligned} E\varphi(j) &= 2 \sum_{\Delta} J(j, j+\Delta) [S(j+\Delta)\varphi(j) \\ &\quad - \{S(j)S(j+\Delta)\}^{1/2} \varphi(j+\Delta)] + E^0\varphi(j), \end{aligned} \quad (23)$$

where $J(j, j+\Delta) = J$ for exchange between host atoms or J' for exchange between a host atom and the impurity atom. The square root term results from the normalization of the impurity state in Eq. (22). We adopt the convention that $j=1$ refers to the impurity site and that $j=2, 3, \dots, z+1$ refers to the z nearest neighbors of the impurity and write

$$\mathbf{U} = \begin{pmatrix} \varphi(1) \\ \varphi(2) \\ \dots \\ \varphi(z+1) \\ \varphi(z+2) \\ \dots \\ \varphi(N) \end{pmatrix}. \quad (24)$$

Equation (23) can then be written as

$$E\mathbf{U} = (\mathbf{H}^0 + \mathbf{H}')\mathbf{U}, \quad (25)$$

where \mathbf{H}^0 is given by Eq. (11) with a shift in the zero-point energy

$$\Delta E^0 = (JS - J'S')SZ + g\beta H_z(S - S'). \quad (26)$$

\mathbf{H}' is a $z+1$ by $z+1$ matrix,

$$\mathbf{H}' = 2JS \begin{pmatrix} \epsilon\sigma & -\gamma & -\gamma & \cdot & \cdot & \cdot & -\gamma \\ -\gamma & \rho & & & & & -\gamma \\ -\gamma & & \rho & & 0 & & \\ \cdot & & & \cdot & & & \\ \cdot & & 0 & & \cdot & & \\ \cdot & & & & & \cdot & \\ -\gamma & & & & & & \rho \end{pmatrix}, \quad (27)$$

where

$$\begin{aligned} \epsilon &= (J' - J)/J, \\ \sigma &= S'/S, \\ \gamma &= \sigma^{1/2}(\epsilon + 1) - 1, \\ \rho &= (\sigma - 1) + \epsilon\sigma. \end{aligned} \quad (28)$$

Equation (25) can formally be solved for the impurity states

$$\begin{aligned} u &= (E - \mathbf{H}^0)^{-1} \mathbf{H}' u, \\ \{I - (E - \mathbf{H}^0)^{-1} \mathbf{H}'\} u &= 0, \end{aligned} \quad (29)$$

where

$$u = \begin{pmatrix} \varphi(1) \\ \varphi(2) \\ \dots \\ \varphi(z+1) \end{pmatrix}, \quad (30)$$

and I is a $z+1$ by $z+1$ unit matrix. The inverse of the matrix $\{I - (E - \mathbf{H}^0)^{-1} \mathbf{H}'\}$ is called the dynamic response function for the system. The energy of the impurity states is determined by the secular determinant

$$\det\{I + \mathbf{G}\mathbf{H}'(2JS)^{-1}\} = 0, \quad (31)$$

where \mathbf{G} is the upper $z+1$ by $z+1$ part of the dimensionless Green's function matrix. The method used in obtaining Eq. (29) is a simple example of Löwdin's partitioning technique.¹⁷ The same result, however, can also be obtained using the Brillouin-Wigner perturbation scheme.

In a simple cubic lattice, the matrix \mathbf{H}' has dimensions 7×7 since a defect atom is connected with itself and its six nearest neighbors. It is not necessary, however, to solve a 7×7 determinantal equation, since the determinant may be factored with the use of a group theory. The wave function for a spin-wave mode associated with a defect must transform according to one of the irreducible representations of the cubic point group. This factorization of the determinant was previously discussed by Koster and Slater⁶ in connection with their consideration of localized electronic states associated with impurities. The problem of determining the proper linear combination of the $\varphi(j)$'s which will lead to

¹⁷ Per-Olov Löwdin, "Studies in Perturbation Theory. IV. Solution of Eigenvalue Problem by Projection Operator Formalism," Quantum Chemistry Group, Uppsala University, Uppsala, Sweden, 1960 (unpublished).

factorization of the secular determinant is exactly analogous to the problem of constructing symmetrized linear combinations of plane waves in an energy band calculation. In the present case, the atoms of interest are located in (0,0,0) and (1,0,0) positions, so that it is possible to form two s -like linear combinations (representation Γ_1),¹⁸ three p -like combinations (Γ_{15}), and two d states (Γ_{12}). In a body-centered cubic lattice, there are two s -like combinations (Γ_1), three p (Γ_{15}), three d ($\Gamma_{25'}$) and one f ($\Gamma_{2'}$). In a face-centered cubic lattice there are, in addition to the same s -, p -, and d -type combinations present for the body-centered cubic lattice, two more d -type combinations (Γ_{12}), and three f -type combinations (Γ_{25}).

III. APPLICATION TO CUBIC CRYSTALS

1. S-Like States for Cubic Crystals

In this section we solve the secular determinant for the s -like states in terms of the diagonal Green's function matrix elements for any cubic crystal. The secular determinant, Eq. (31), will be of order 7, 9, or 13 for simple cubic, body-centered cubic and face-centered cubic crystals, respectively. The solution of this determinant is greatly facilitated by transforming to the representation determined by the basis functions for the irreducible representations of the cubic point group. In this representation the secular determinant will factor into subdeterminants; one subdeterminant for each irreducible representation contained in the reducible representation defined by the $\varphi(j)$'s. The low-lying states are the s -like states which correspond to the eigenvectors

$$u_{s1} = \begin{pmatrix} 1 \\ 0 \\ 0 \\ \vdots \\ 0 \end{pmatrix}, \quad u_{s2} = z^{-1/2} \begin{pmatrix} 0 \\ 1 \\ 1 \\ 1 \\ \vdots \\ 1 \end{pmatrix}, \quad (32)$$

where the impurity is at R_1 and its nearest neighbors are located R_2, R_3, \dots, R_{z+1} . The secular subdeterminant for the s -like states in the representation defined by u_{s1} and u_{s2} is

$$\det \left\{ I + \frac{1}{2} \begin{pmatrix} G_{11}, & z^{1/2} G_{12} \\ z^{1/2} G_{12}, & \sum_{z=2}^{z+1} G_{i2} \end{pmatrix} \begin{pmatrix} z\epsilon, & -z^{1/2}\gamma \\ -z^{1/2}\gamma, & \rho \end{pmatrix} \right\} = 0, \quad (33)$$

where the G_{ij} are the matrix elements of the dimensionless Green's function on the basis of the functions in (32). Another simplification which results for cubic crystals is that all of the Green's function matrix elements in Eq. (33) can be expressed in terms of the diagonal element G_{11} . For example, taking the origin at the impurity site

$$\begin{aligned} G_{12} &= N^{-1} \sum_q \exp\{i\mathbf{q} \cdot \mathbf{R}(2)\} (\mathcal{E} - \mathcal{E}_q)^{-1}, \\ &= 2(NZ)^{-1} \sum_q \mathcal{E}_q (\mathcal{E} - \mathcal{E}_q)^{-1}, \\ &= 2z^{-1} (\mathcal{E} G_{11} - 1). \end{aligned} \quad (34)$$

It is important to note that the exact value of G_{12} at $\mathcal{E}=0$ is $-2/z$. Similarly,

$$\begin{aligned} \sum_{i=2}^{z+1} G_{i2} &= N^{-1} \sum_q \exp\{-i\mathbf{q} \cdot \mathbf{R}(2)\} \\ &\quad \times \left[\sum_{\Delta} \exp\{i\mathbf{q} \cdot \mathbf{R}(\Delta)\} \right] (\mathcal{E} - \mathcal{E}_q)^{-1} \\ &= 4z^{-1} \sum_q \mathcal{E}_q^2 (\mathcal{E} - \mathcal{E}_q)^{-1} = 2\mathcal{E} G_{12}, \end{aligned} \quad (35)$$

where we have used the fact that $\sum_q \mathcal{E}_q = 0$. Furthermore, in the limit as N becomes very large

$$G_{11} = N^{-1} \sum_q (\mathcal{E} - \mathcal{E}_q)^{-1} = \Omega^{-1} \int dq (\mathcal{E} - \mathcal{E}_q)^{-1}, \quad (36)$$

where the integral is over the Brillouin zone and Ω is the volume of the Brillouin zone. We may now write G_{11} in terms of the density of states $n(\mathcal{E})$,

$$G_{11} = \int_0^{\mathcal{E}_m} d\mathcal{E}' (\mathcal{E} - \mathcal{E}')^{-1} n(\mathcal{E}'), \quad (37)$$

where \mathcal{E}_m is the maximum spin-wave energy. Thus, we have the important result that the s -like states are completely determined by the density of states. For a given $\epsilon = (J' - J)/J$ and $\sigma = S'/S$ Eq. (33) is a transcendental equation determining the energy. Alternately we may choose an energy and value of σ and regard Eq. (33) as determining the value of ϵ . Using the definitions of γ and ρ we may write

$$\begin{pmatrix} z\epsilon & -z^{1/2}\gamma \\ -z^{1/2}\gamma & \rho \end{pmatrix} = \epsilon \begin{pmatrix} z & -(z\sigma)^{1/2} \\ -(z\sigma)^{1/2} & \sigma \end{pmatrix} + \begin{pmatrix} 0 & 1 - \sigma^{1/2} \\ 1 - \sigma^{1/2} & \sigma - 1 \end{pmatrix}. \quad (38)$$

¹⁸ The states we have designated s , p , d , and f are more properly labeled by the appropriated irreducible representations of the cubic group. These are given by L. P. Bouckaert, R. Smoluchowski, and E. P. Wigner, Phys. Rev. **50**, 58 (1936).

The important point here is that the matrix multiplying ϵ is singular. This means that the secular determinant, Eq. (33), is linear in ϵ . Solving for ϵ we have

$$\epsilon = [-1 + (1 - \sigma)G_{12}(\mathcal{E} - \frac{1}{2}z)] \times [z/2G_{11} + G_{12}\{(\sigma - 1)(\mathcal{E} - \frac{1}{2}z) + (\mathcal{E} - z)\}], \quad (39)$$

which is the condition for the existence of a localized s -like state. Virtual states, as we shall see, can result when the real part of the equation is satisfied.

2. Simple Cubic Crystal

The case of the simple cubic crystal is particularly important from a theoretical point of view because relatively simple expressions can be obtained for the matrix elements of the Green's function. Although there are no real ferromagnetic crystals with this magnetic structure, it is reasonable to expect that the qualitative features of the impurity problem do not depend critically upon the crystal structure.

We proceed according to the general theory outlined in Sec. IV. The secular determinant for the seven impurity states, Eq. (31), is block diagonalized by unitary transformation V :

$$V = \begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & a & b & 0 & 0 & 0 & d \\ 0 & a & -b & 0 & 0 & 0 & d \\ 0 & a & 0 & b & 0 & c & e \\ 0 & a & 0 & -b & 0 & c & e \\ 0 & a & 0 & 0 & b & -c & e \\ 0 & a & 0 & 0 & -b & -c & e \end{pmatrix}, \quad \begin{aligned} a &= (1/6)^{1/2}, \\ b &= (1/2)^{1/2}, \\ c &= 1/2, \\ d &= (1/3)^{1/2}, \\ e &= (1/12)^{1/2}, \end{aligned} \quad (40)$$

where the atoms are located by the vectors $\mathbf{R}(1) = (0, 0, 0)$, $\mathbf{R}(2) = (1, 0, 0)$, $\mathbf{R}(3) = (-1, 0, 0)$, $\mathbf{R}(4) = (0, 1, 0)$, $\mathbf{R}(5) = (0, -1, 0)$, $\mathbf{R}(6) = (0, 0, 1)$, and $\mathbf{R}(7) = (0, 0, -1)$. The first two columns of V are the symmetry eigenvector for the s -like states which transform according to the Γ_1 irreducible representation of the full cubic group O_h . The next three columns are p -like states which transform according to the Γ_{15} irreducible representation, and the last two columns are d -like states and transform according to Γ_{12} . One has

$$\det\{V^\dagger[I + \frac{1}{2}GH'(2JS)^{-1}]V\} = [\det A][1 + \frac{1}{2}\rho(G_{11} - G_{23})]^3 \times [1 + \frac{1}{2}\rho(G_{11} + G_{23} - 2G_{24})]^2 = 0, \quad (41)$$

where $\det A$ is given by Eq. (33) with $z=6$ and determines the energy of the s states which according to Eq. (39) is given by the condition that

$$\epsilon = [-1 + (1 - \sigma)G_{12}(\mathcal{E} - 3)] \times [3G_{11} + G_{12}\{(\sigma - 1)(\mathcal{E} - 3) + (\mathcal{E} - 6)\}]. \quad (42)$$

Whenever the second term of Eq. (41) vanishes p states exist, while d states exist when the third term vanishes. The matrix elements G_{ij} are given formally by Eq. (21)

and may be written as integrals in the limit of large N

$$G_{ij} = \Omega^{-1} \int \exp[i\mathbf{q} \cdot \{\mathbf{R}(i) - \mathbf{R}(j)\}] (\mathcal{E} - \mathcal{E}_q)^{-1} d^3q. \quad (43)$$

In the case of a simple cubic lattice, the integrals over the Brillouin zone may be reduced to a single one-dimensional integral involving a product of three Bessel functions and an exponential or trigonometric function. This reduction has been discussed by Koster and Slater.⁶ There are, however, some minor errors in their formulas, and we have found it desirable to summarize the results concerning these integrals in the appendix. The results are that for $\mathbf{R}(i) - \mathbf{R}(j) = (k, m, n)$ we have the integral representation

$$G_{ij}(\mathcal{E}) = i^{(k+m+n+1)} \int_0^\infty dt e^{-t\mathcal{E}} J_k(t) J_m(t) J_n(t), \quad (44)$$

where the J 's are Bessel functions of integral order with real argument t . In terms of the parameter \mathcal{E} the top of the spin-wave band corresponds to $\mathcal{E} = -3$ and the bottom of the band to $\mathcal{E} = +3$. It is clear from Eq. (44) that the real and imaginary components of G_{ij} are either symmetric or antisymmetric in \mathcal{E} and, therefore, we need only calculate the values of the G_{ij} for $\mathcal{E} > 0$. For $|\mathcal{E}| > 3$ the Green's function is real and we may use the integral representation given by Koster and Slater⁶

$$G_{ij}(\mathcal{E}) = \int_0^\infty dt e^{-\mathcal{E}t} I_k(t) I_m(t) I_n(t), \quad \mathcal{E} \geq 3, \quad (45)$$

where the I 's are Bessel functions of integral order and imaginary argument.

Some of these Green's function integrals have been calculated by Koster and Slater⁶ for energies outside of the continuum. Since it was desired to investigate the virtual states as well as the localized states the integrals, Eq. (44), were calculated both outside and inside of the continuum. The results are tabulated in Table I. The numerical integration was carried out on the IBM 7090 electronic computer using Simpson's rule with an interval of 0.01 from 0 to 50. The tabulated values were cross checked using the relationships between the Green's function matrix elements given in the Appendix. The values obtained are in agreement with those calculated by Koster and Slater.⁶ The estimated accuracy of the numerical integration is about 5%. The equation for the existence of localized modes, Eq. (41), has solutions only when the energy of the defect state is above or below the spin-wave band. Resonances or virtual states may, however, exist in the band. From a scattering point of view the virtual states correspond to maxima in the scattering cross section. It is also possible to have bound impurity states below the bottom of the spin-wave band. It turns out that it is necessary to have $J' < 0$ in order to obtain a state lying below the band

TABLE I. The values of several simple cubic integrals. $C(k,m,n) = \int_0^\infty J_k(t)J_m(t)J_n(t) \cos(\varepsilon t)dt$,

$$S(k,m,n) = \int_0^\infty J_k(t)J_m(t)J_n(t) \sin(\varepsilon t)dt.$$

ε	$S(000)$	$C(000)$	$C(100)$	$S(100)$	$S(110)$	$C(110)$	$S(200)$	$C(200)$
0.0	0.0000	0.8954	0.3333	0.0000	0.0000	0.1855	0.0000	0.1542
0.2	0.0740	0.8978	0.3284	0.0599	0.0369	0.1833	0.0580	0.1400
0.4	0.1530	0.8967	0.3129	0.1195	0.0733	0.1745	0.1096	0.1041
0.6	0.2416	0.8999	0.2850	0.1800	0.1116	0.1610	0.1381	0.0408
0.8	0.3557	0.9071	0.2384	0.2419	0.1508	0.1417	0.1333	-0.0480
1.0	0.6027	0.8708	0.1324	0.2903	0.2268	0.1032	-0.0391	-0.1218
1.2	0.6268	0.6171	0.0826	0.2468	0.2109	-0.0014	-0.0188	0.0295
1.4	0.6057	0.5063	0.0507	0.2363	0.1803	-0.0489	0.0263	0.0406
1.5	0.5988	0.4631	0.0339	0.2316	0.1642	-0.0673	0.0440	0.0376
1.6	0.5909	0.4259	0.0182	0.2271	0.1481	-0.0828	0.0565	0.0307
1.8	0.5762	0.3602	-0.0124	0.2161	0.1151	-0.1062	0.0714	0.0063
2.0	0.5621	0.3040	-0.0414	0.2027	0.0803	-0.1213	0.0756	-0.0209
2.2	0.5496	0.2537	-0.0697	0.1860	0.0444	-0.1291	0.0651	-0.0479
2.4	0.5378	0.2055	-0.0967	0.1644	0.0076	-0.1285	0.0446	-0.0713
2.5	0.5321	0.1826	-0.1108	0.1522	-0.0114	-0.1244	0.0299	-0.0788
2.6	0.5260	0.1585	-0.1225	0.1374	-0.0309	-0.1175	0.0116	-0.0834
2.8	0.5167	0.1062	-0.1447	0.0991	-0.0703	-0.0929	-0.0322	-0.0804
3.0	0.4926	0.0	-0.1593	0.0	-0.0977	-0.1269	-0.0727	0.0
3.1	0.4311	0.0	-0.1121	0.0	-0.0568	0.0	-0.0376	0.0
3.2	0.4001	0.0	-0.0934	0.0	-0.0428	0.0	-0.0269	0.0
3.5	0.3410	0.0	-0.0645	0.0	-0.0242	0.0	-0.0141	0.0
4.2	0.2646	0.0	-0.0371	0.0	-0.0137	0.0	-0.0056	0.0
5.0	0.2142	0.0	-0.0237	0.0	-0.0053	0.0	-0.0028	0.0
5.5	0.1897	0.0	-0.0144	0.0	-0.0038	0.0	-0.0020	0.0
6.0	0.1725	0.0	-0.0127	0.0	-0.0028	0.0	-0.0014	0.0

($\varepsilon > 3$); this situation corresponds to an impurity which is coupled antiferromagnetically to the remainder of the ferromagnetic system. It is obvious that such a configuration would be unstable, so we further limit our detailed considerations to the case $J' > 0$. For a fixed value of the ratio of spins σ and exchange integrals J'/J , the p and d states have considerably higher energy than the s state. Solutions of Eq. (41) are shown in Fig. 1 for three values of the spin ratio. If the spin of the defect atom is less than or equal to that of the host ($\sigma \leq 1$), it

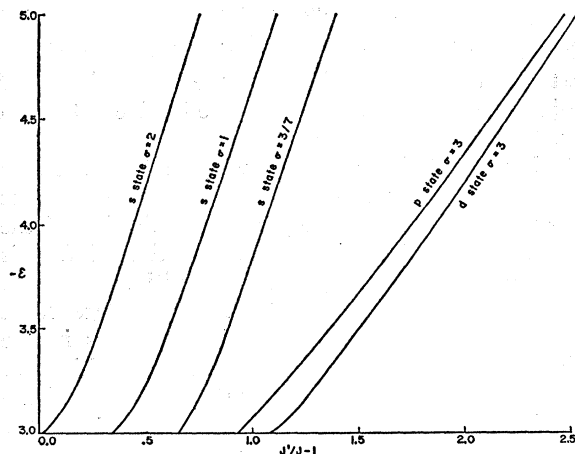


FIG. 1. The energies of localized spin-wave modes are shown for several values of the spin ratio $\sigma = S'/S$ as a function of $J'/J-1$. The dimensionless energy $-\varepsilon$, which is positive, is related to the "true" energy E by $-\varepsilon = (E/4JS) - 3$.

is necessary to have $J'/J > 1$ (at $\sigma = 1$, $J'/J = 1.34$ for $\varepsilon = -3$) to obtain a localized mode. If $0 \leq J'/J \leq 1$ resonant states may exist: these will be discussed in the next section. However, for sufficiently large $\sigma (\geq 2.05)$, localized states may exist with $J'/J < 1$.

IV. RESONANT STATES, GENERAL

If the energy ε of the state considered lies in the spin-wave band, the integrand of Eq. (21) is singular, due to the presence of the quantity $\varepsilon - \varepsilon_q$ in the denominator. It is then necessary to specify how the integral is to be defined. We follow the usual procedure according to which the energy is regarded as having a small, positive, imaginary part. With this definition, the matrix elements G_{ij} are complex. It is not generally possible to satisfy Eq. (41) for real ε ; however, these equations may be satisfied by the real parts of the complex Green's function. Under certain conditions, which will become apparent in the course of this discussion, a "virtual state" or "resonance" may exist when the real part of the secular determinant vanishes.

The situation can be investigated in detail by determining the contribution of the impurity states to the density of states. The density of spin-wave states may be written formally as¹⁹

$$n(E) = \frac{-1}{\pi} \text{Im} \left(\text{Tr} \frac{1}{E^+ - \mathbf{H}} \right), \quad (46)$$

¹⁹ J. R. Klauder, Ann. Phys. (N. Y.) 14, 43 (1961).

where H is the full spin-wave Hamiltonian for the system including the defect and Tr denotes the trace. We write E^+ to indicate that the energy may be regarded as having an infinitesimal, positive, imaginary part. This equation may be rewritten as

$$\begin{aligned} n(E) &= \frac{-1}{\pi} \text{Im} \left[\text{Tr} \frac{d}{dE} \{ \ln(E^+ - H) \} \right] \\ &= \frac{-1}{\pi} \text{Im} \left[\frac{d}{dE} \ln \{ \det(E^+ - H) \} \right]. \end{aligned} \quad (47)$$

To separate the contribution from the impurity states, we write

$$(E^+ - \mathbf{H}) = (E^+ - \mathbf{H}^0) \left(1 - \frac{1}{E^+ - \mathbf{H}^0} \mathbf{H}' \right), \quad (48)$$

and use the fact that the determinant of the product of two matrices is equal to the product of the determinants. In addition, the density of states for the system with the impurities replaced by normal atoms is

$$\begin{aligned} n^0(E) &= \frac{-1}{\pi} \text{Im} \left(\text{Tr} \frac{1}{E^+ - \mathbf{H}^0} \right) \\ &= \frac{-1}{\pi} \text{Im} \left[\frac{d}{dE} \ln \{ \det(E^+ - \mathbf{H}^0) \} \right]. \end{aligned} \quad (49)$$

The contribution of the impurities to the density of states is, therefore,

$$\begin{aligned} \Delta n &= n - n^0 \\ &= \frac{-1}{\pi} \text{Im} \left[\frac{d}{dE} \ln \{ \det(I - (E^+ - \mathbf{H}^0)^{-1} \mathbf{H}') \} \right]. \end{aligned} \quad (50)$$

Equation (50) is exact. The localized modes previously studied were obtained by requiring that the determinant of the matrix representing the operator $\{1 - [1/(E^+ - \mathbf{H}^0)]\mathbf{H}'\}$ must vanish. We were able to factor the determinant into a product of factors, and this factorization is still valid, although the determinant does not vanish. The factors of the determinant will be denoted simply u_j .

$$\det\{I - (E^+ - \mathbf{H}^0)^{-1} \mathbf{H}'\} = \pi_j u_j, \quad (51)$$

$$\begin{aligned} \Delta n &= \frac{-1}{\pi} \sum_j \text{Im} \left(\frac{d}{dE} \ln u_j \right), \\ &= \frac{-1}{\pi} \sum_j \text{Im} \left(u_j^{-1} \frac{du_j}{dE} \right), \end{aligned} \quad (52)$$

and the contribution to the density of states from a particular impurity state is just

$$\Delta n_j = \frac{-1}{\pi} \text{Im} \left(u_j^{-1} \frac{du_j}{dE} \right). \quad (53)$$

The behavior of this quantity near a resonance is to be investigated. The real and imaginary parts of u_j are denoted by $u_{j,r}$ and $u_{j,i}$, respectively. The resonant state will be said to be located at the energy, E_r , for which the real part of u_j vanishes. This will not, in general, be the actual maximum of the density of states which will be shifted slightly depending on $u_{j,i}$. This definition of the resonant energy is, however, the conventional one. We assume that both the energy derivative of $u_{j,r}$ and the imaginary part of u_j are not zero at E_r . If the resonance does not occur at a critical point in the density of states we may expand u_j near E_r :

$$u_j(E) = (E - E_r)u_{j,r}'(E_r) + iu_{j,i}(E), \quad (54)$$

where the prime denotes differentiation with respect to energy. Then, near E_r

$$\Delta n_j = \frac{1}{\pi} \frac{\{ (u_{j,i}/u_{j,r}') - (E - E_r)(u_{j,i}'/u_{j,r}') \}}{(E - E_r)^2 + (u_{j,i}/u_{j,r}')^2}. \quad (55)$$

In particular, for $E = E_r$,

$$\Delta n_j(E_r) = \frac{1}{\pi} (u_{j,r}'/u_{j,i}).$$

Equation (61) may be used to define a level width. If we use (49) to specify the contribution to the density of states from a single level of complex energy $E_0 - i\Gamma$ (Γ positive), we find

$$n(E) = \frac{1}{\pi} \frac{\Gamma}{(E - E_0)^2 + \Gamma^2}, \quad (57)$$

$$n(E_0) = 1/\pi\Gamma.$$

Comparison of Eqs. (56) and (57) leads to the following general definition of the level width, Γ :

$$\Gamma = (u_{j,i}/u_{j,r}'). \quad (58)$$

The level width is then the ratio of the imaginary part of the contribution to the determinant of $1 - [1/(E^+ - \mathbf{H}^0)]\mathbf{H}'$ to the energy derivative of the real part for a particular state.

This result is in agreement with that deduced by Brout and Visscher³ from consideration of the dynamic response function. However, the algebraic sign of the ratio $u_{j,i}/u_{j,r}'$ arising in the computation of (58) is significant. It is necessary that Γ be positive in order to have a virtual or resonant state, since such states must produce an increase of the density of states in the neighborhood. The requirement that Γ be positive can be used to distinguish resonant states from other situations in which the real part of u_j may vanish. The other cases (negative Γ) correspond to regions in which the density of states is diminished. Since the total number of states in the system is unaffected by the interaction \mathbf{H}' (no atoms have been added or removed), an increase

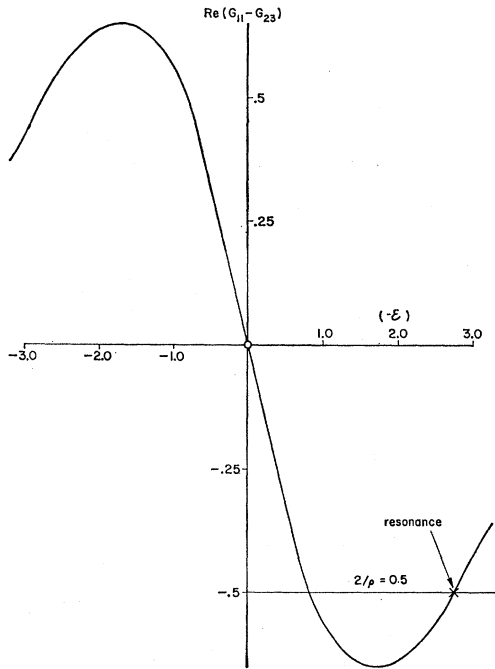


FIG. 2. The real part of $y = (G_{11} - G_{23})$ is plotted as a function of the dimensionless energy, $-\epsilon$. The spin-wave continuum lies between $-3 \leq \epsilon \leq 3$. Resonant states are located by determining the intersection of this curve with the straight line $y = 2/\rho = \text{const}$. Only those intersections for which the derivative of $(G_{11} - G_{23})$ (with respect to the true spin-wave energy) is positive represent resonant states.

of the density of states in some region of energy implies a corresponding decrease in another. The latter do not, however, correspond to resonances.

V. LOCATIONS AND WIDTHS OF RESONANT SPIN-WAVE STATES

The formalism developed in the preceding section to enable the calculation of the width of a resonant state may be readily applied to the spin-wave problem. The equations derived in Sec. III from the factored secular determinant may be used directly. Expressions for both the real and imaginary parts of the complex integrals G_{ij} are given in the Appendix.

Possible locations of resonant states are found by requiring the real part of the secular determinant to vanish, while the width is computed from Eq. (58). The determination of the location and widths of the states is illustrated graphically in Fig. 2 for the p state. The quantity $G_{11} - G_{23}$ is plotted as a function of energy. The possible resonant states occur at the intersection of the curve representing $G_{11} - G_{23}$ with the straight line representing $2/|\rho|$. When $|\rho|$ is smaller than 3.08 no intersection is possible, and neither localized states nor resonances exist. For $|\rho|$ greater than this, two intersections of the curve with the straight line will occur, however only one of these indicates a resonance, since only one

has a positive Γ . This follows since (58) yields in this case

$$\Gamma = \text{Im}(G_{11} - G_{23}) / \text{Re}(G_{11}' - G_{23}'). \quad (59)$$

The quantity $\text{Im}(G_{11} - G_{23})$ is always negative, so that a positive Γ will occur only if $\text{Re}(G_{11}' - G_{23}')$ is also negative.

The locations and widths of resonant states are shown in Fig. 3 for certain values of the spin ratio $\sigma = S'/S$. We note that reasonably long-lived virtual states can exist near the top ($\mathcal{E} = -3$) or bottom ($\mathcal{E} = 3$) of the spin-wave band.

VI. DISCUSSION AND CONCLUSION

In the preceding sections we have developed the theory of spin-wave impurity states in ferromagnets. We have established that localized impurity states can exist for reasonable values of the ratios of the impurity spin and exchange to those of the host crystal. It has also been noted that localized states below the spin-wave band occur when the impurity atom is coupled antiferromagnetically to the host spins. This configuration will be unstable since the state in which the impurity spin is antiparallel to the host spins has lower energy. The formalism developed here may still be employed provided the vacuum for the perturbed ferromagnet is taken with the impurity antiferromagnetically aligned.

The nature of the low-lying s states for any cubic crystal has been investigated. A formula, Eq. (39), relating the low-lying impurity state energy to the change in spin and exchange has been given which depends only on the density of states. The impurity states for a simple cubic crystal have been investigated in detail.

The localized states with energy above the continuum are stationary states with infinite lifetimes (neglecting magnon-magnon interactions). Virtual states which decay into the continuum result when the real part of the

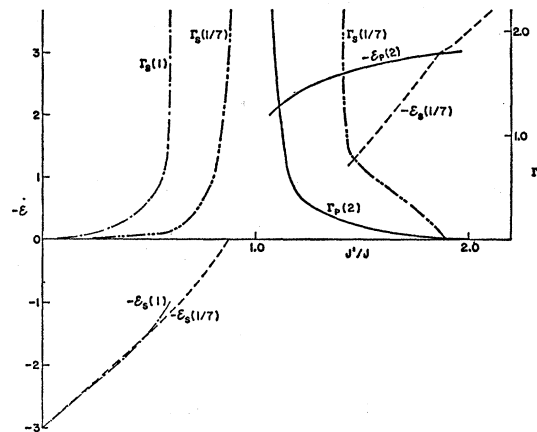


FIG. 3. The energies ($-\epsilon$) and widths (Γ) of some localized spin-wave states are shown as functions of the exchange integral ratio J'/J for some values of the spin ratio $\sigma = S'/S$. Note different scales for the energy and the width.

secular determinant vanishes. By considering the change in the density of states due to impurities it was shown that only virtual states with positive width correspond to real resonances. An expression for the width of a resonant state has been given in terms of the real and imaginary parts of the secular determinant. Virtual states with long lifetimes can be formed near the bottom of the spin-wave band. These states are particularly important and may cause significant effects in the physical properties of impure ferroinsulators since they are excited at low temperatures.

We have treated numerically only the case of a simple cubic crystal, whereas ferroinsulators such as the rare-earth binary compounds have the magnetic atoms arranged in a face-centered cubic configuration. The qualitative results concerning the conditions under which it is possible to form impurity states will not depend critically upon the crystal structure.

It may be possible to observe the spin-wave impurity states by ferromagnetic resonance in the far infrared. Virtual states may make an observable contribution to the thermal resistance in the spin-wave conductivity and to the spin-wave specific heat at low temperatures. The effect of impurities on these quantities is presently being considered.

Impurity states in antiferromagnets or ferrimagnets should have some additional interesting features because of the complexity of the density of states. Furthermore, in these crystals the density of states can be altered by an external magnetic field. In the ferromagnet an external magnetic field causes no change in the shape of the density of states, but simply shifts the entire curve to higher energies.

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APPENDIX: INTEGRAL REPRESENTATIONS

1. Green's Function for a Simple Cubic Crystal

Integral representations for the Green's function for a simple cubic lattice have been discussed by Koster and Slater,⁶ however, since the formula (A8) in their paper is incorrect, an alternate derivation is presented in this Appendix.

The matrix elements of the dimensionless Green's function are given by

$$G_{ij} = N^{-1} \sum_q \exp[i\mathbf{q} \cdot \{\mathbf{R}(i) - \mathbf{R}(j)\}] (\mathcal{E} - \mathcal{E}_q)^{-1}, \quad (\text{A1})$$

where

$$\mathcal{E}_q = \frac{1}{2} \sum_{\Delta} \cos\{\mathbf{q} \cdot \mathbf{R}(\Delta)\}. \quad (\text{A2})$$

The sum over Δ is a sum over the z vectors $\mathbf{R}(\Delta)$ which connect an atom with its z nearest neighbors. Writing $\mathbf{R}(i) - \mathbf{R}(j) = (k, m, n)$

$$G_{ij} = \frac{1}{N} \sum_q \frac{\exp\{i(q_x k + q_y m + q_z n)\}}{\mathcal{E} - (\cos q_x + \cos q_y + \cos q_z)}. \quad (\text{A3})$$

In the limit of large N we may replace the sum over the propagation vectors by an integral over the first Brillouin zone.

$$G_{ij} = \frac{1}{\Omega} \int d\mathbf{q} \frac{\exp\{i(q_x k + q_y m + q_z n)\}}{n - (\cos q_x + \cos q_y + \cos q_z)}. \quad (\text{A4})$$

We wish to derive an integral representation for G_{ij} which is valid both inside and outside of the spin wave continuum. We, therefore, allow \mathcal{E} to be displaced slightly off of the real axis so that the denominator is never zero.²⁰

$$\begin{aligned} G_{ij}(\mathcal{E} - is) &= \frac{1}{(2\pi)^3} \int d\mathbf{q} \frac{\exp\{i(q_x k + q_y m + q_z n)\}}{\mathcal{E} - is - (\cos q_x + \cos q_y + \cos q_z)} \\ &= \frac{i}{(2\pi)^3} \int_0^\infty dt \exp -i(\mathcal{E} - is)t \\ &\quad \times \int_{-\pi}^{\pi} dq_x \exp i(q_x k + t \cos q_x) \\ &\quad \times \int_{-\pi}^{\pi} dq_y \exp i(q_y m + t \cos q_y) \\ &\quad \times \int_{-\pi}^{\pi} dq_z \exp i(q_z n + t \cos q_z). \quad (\text{A5}) \end{aligned}$$

Using the integral representation for the Bessel function²¹

$$J_p(t) = \frac{i^{-p}}{2\pi} \int_{-\pi}^{\pi} \exp i(p\xi + t \cos \xi) d\xi, \quad (\text{A6})$$

and letting $s \rightarrow 0$

$$\begin{aligned} \lim_{s \rightarrow 0} G_{ij}(\mathcal{E} - is) \\ = i^{(k+m+n+1)} \int_0^\infty dt e^{-is t} J_k(t) J_m(t) J_n(t). \quad (\text{A7}) \end{aligned}$$

Outside of the continuum, $\mathcal{E} > 3$ it can easily be shown that the imaginary part of G vanishes and one may use the representation given by Koster and Slater⁶

$$G_{ij}(\mathcal{E}) = \int_0^\infty dt e^{-\mathcal{E}t} I_k(t) I_m(t) I_n(t), \quad (\text{A8})$$

where I 's are the Bessel functions of imaginary argument.

²⁰ Since \mathcal{E} is proportional to the negative of the real spin-wave energy we take the imaginary part to be negative. This is equivalent to letting E have a small positive imaginary part.

²¹ See, for example, J. A. Stratton, *Electromagnetic Theory* (McGraw-Hill Book Company, Inc., New York, 1941), p. 367.

2. Interdependence of Matrix Elements

The relationships between the matrix elements of the Green's function derived in Sec. IV can be obtained from the integral representation using integration by parts. For example,

$$G_{11} = \int_0^\infty dt e^{-i\mathcal{E}t} J_0(t)^3 = \frac{1}{\mathcal{E}} \left(-3 \int_0^\infty dt e^{-i\mathcal{E}t} J_0(t)^2 J_1(t) + 1 \right), \quad (A9)$$

which agrees with Eq. (38). Similarly,

$$G_{12} = - \int_0^\infty dt e^{-i\mathcal{E}t} J_0(t)^2 J_1(t) = \frac{1}{\mathcal{E}} \left[-2i \int_0^\infty dt e^{-i\mathcal{E}t} J_1(t)^2 J_0(t) + \frac{1}{2} \int_0^\infty dt e^{-i\mathcal{E}t} J_0(t)^3 - \frac{i}{2} \int_0^\infty dt e^{-i\mathcal{E}t} J_0(t)^2 J_2(t) \right], \quad (A10)$$

which is the same as Eq. (39). Other relations between the matrix elements of the Green's function can also be obtained. These relationships are valuable checks on the accuracy of any numerical evaluation of the Green's function.

3. Density of States

Using the well-known result that

$$\lim_{\epsilon \rightarrow 0} \frac{1}{x - i\epsilon - y} = \mathcal{P} \frac{1}{x - y} + i\pi \delta(x - y), \quad (A11)$$

where \mathcal{P} denotes the principal part, it follows from (A4) that

$$\text{Im} G_{11}(\mathcal{E}) = \pi n(\mathcal{E}), \quad (A12)$$

where Im denotes the imaginary part and $n(\mathcal{E})$ is the density of states. Thus, for the simple cubic lattice we have the well-known result²² that

$$n(\mathcal{E}) = \frac{1}{\pi} \int_0^\infty dt \cos(\mathcal{E}t) J_0(t)^3. \quad (A13)$$

This expression can be obtained directly from the definition of the density of states

$$n(\mathcal{E}) = \frac{1}{(2\pi)^3} \int d\mathbf{q} \delta(\mathcal{E} - \mathcal{E}_q). \quad (A14)$$

Introducing the integral representation of the delta function

$$\delta(\mathcal{E} - \mathcal{E}_q) = \frac{1}{2\pi} \int_{-\infty}^\infty dt \exp\{i(\mathcal{E} - \mathcal{E}_q)t\} \quad (A15)$$

one has

$$n(\mathcal{E}) = \frac{1}{2\pi} \int_{-\infty}^\infty dt e^{-i\mathcal{E}t} \left(\frac{1}{2\pi} \int_{-\pi}^\pi e^{it \cos q} dq \right)^3 = \frac{1}{2\pi} \int_{-\infty}^\infty dt e^{-i\mathcal{E}t} J_0(t)^3 = \frac{1}{\pi} \int_0^\infty dt \cos(\mathcal{E}t) J_0(t)^3. \quad (A16)$$

²² E. W. Montroll, in *Proceedings of the Third Berkeley Symposium on Mathematical Statistics and Probability* (University of California Press, Berkeley, California, 1956), Vol. 3, p. 209.