PHYSICAL REVIEW VOLUME 167, NUMBER 2 10 MARCH 1968

Coupling between Impurity Spins in a Magnetic Host*

ROBERT M. WHITE AND C. MICHAEL HOGAN

Department of Physics, Stanford University, Stanford, California

(Received 11 October 1967; revised manuscript received 29 November 1967)

The presence of an impurity spin in a Heisenberg ferromagnet can give rise to localized modes whose energies may lie above or within the spin-wave band, depending upon the magnitude of the impurity spin and its coupling to the host. We have calculated the eigenvalues of the *p*-like modes for a one-dimensional chain and a simple cubic lattice when two such impurities are present. In the case where the single-impurity mode lies outside the spin-wave band, we find two modes symmetrically located with respect to this original mode. This mode splitting defines an effective impurity-impurity interaction. We find that this interaction can be quite large when the modes lie close to the spin-wave band. When the single-impurity mode lies within the band, it is characterized by a redistribution of the density of states. We have also investigated this redistribution for the case of two impurities. It is found that these results are considerably different from those obtained by a spin-wave scattering approach. This difference illustrates the importance of the contribution of bound states to indirect coupling.

I. INTRODUCTION

THE localized modes associated with impurities in crystals have been the object of many recent investigations. For example, localized excitons have been found to play an important role in the fluorescence of optically active materials. Similarly, localized phonons are a useful probe for studying lattice dynamics. In this paper we shall be concerned with spin impurities in a magnetic host and their associated localized modes. Although this problem has had a long history of study within the framework of molecular-field theory, the first exact treatment of localized magnon modes at zero temperature was made by Wolfram and Callaway.[1] Using Green's-functions techniques they considered the conditions for the existence of the localized modes of a ferromagnetically coupled impurity in a ferromagnetic host. Ishii, Kanamori, and Nakamura[2] used spin-wave techniques to investigate the modes associated with an antiferromagnetically coupled impurity in a ferromagnet. This case has also been investigated recently by Wang and Callen.[3] The case of a spin impurity in an antiferromagnetic host has been examined by Tonegawa and Kanamori.[4] All these results were obtained for zero temperature. The thermodynamic properties of such impurity modes have been studied by Hone, Callen, and Walker.[5]

In all these calculations the interactions between impurities were neglected. At first, this might appear reasonable if the concentration is not large. However, it is known that nuclear spins in ferro and antiferromagnetic hosts are coupled by the virtual exchange of spin waves.[6,7] This so-called Suhl-Nakamura interaction is predicted to extend over many lattice spacings. The purpose of this present paper is to investigate whether such coupling might exist between localized magnon modes.

In order to resolve this question, we have considered the problem of two impurities in a linear chain and in a simple cubic lattice. We used the Green's-function approach of Wolfram and Callaway, which is reviewed in Sec. II, with application to a single impurity in a linear chain, as shown schematically in Fig. 1. In Sec. III we add a second impurity and determine how the single-impurity states are split. This splitting is taken as a measure of the effective impurity-impurity coupling. In particular, consider two spins $\mathbf{S}_1$ and $\mathbf{S}_2$ ($|\mathbf{S}_2|=|\mathbf{S}_1|=S$). In the presence of an external field $\mathbf{H}$, and the absence of any coupling between the spins, the first excited state of the pair is twofold degenerate, with an energy $g\mu_B H$. If a coupling of the form $J\mathbf{S}_1\cdot\mathbf{S}_2$ is introduced, this first excited state splits by an amount $2JS$. Thus, the localized-mode splitting divided by twice the magnitude of the impurity spin gives the effective coupling J. We find that when the local modes lie outside the spin-wave continuum, their interaction depends upon the overlap of the localized mode wave functions. Thus, if the modes have energies far above the spin-wave band and are very localized, the interaction is very small. On the other hand, if the modes are close to the spin-wave band, their interaction will be appreciable. In Sec. IV these results are com-

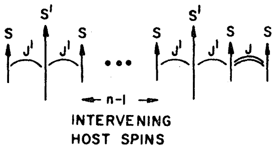

FIG. 1. Linear-chain geometry for impurity-impurity coupling.

---

* Supported by the Advanced Research Projects Agency through the Center for Materials Research at Stanford University Stanford, Calif.

[1] T. Wolfram and J. Callaway, Phys. Rev. 130, 2207 (1963).

[2] H. Ishii, J. Kanamori, and T. Nakamura, Progr. Theoret. Phys. (Kyoto) 33, 795 (1965).

[3] Y. Wang and H. Callen, Phys. Rev. 160, 358 (1967).

[4] T. Tonegawa and J. Kanamori, Phys. Letters 21, 130 (1966).

[5] D. Hone, H. Callen, and L. Walker, Phys. Rev. 144, 283 (1966).

[6] T. Nakamura, Progr. Theoret. Phys. (Kyoto) 20, 542 (1958).

[7] H. Suhl, J. Phys. Radium 20, 333 (1959).

pared with those obtained from a perturbation calculation. We find that when the impurity is sufficiently different from the host to produce a local mode, the neglect of bound states in the perturbation approach can lead to considerably different results. In Sec. V we present computer results for the simple cubic lattice. These are qualitatively the same results as those of the linear chain. However, the linear chain has the advantage that the results may be expressed in a relatively simple analytic form.

II. EIGENVALUES FOR ONE IMPURITY

In the absence of an impurity the isotropic exchange Hamiltonian for a linear chain of spins is

$$\mathbf{H}_0 = -J\sum_n \sum_{\delta=\pm 1} \mathbf{S}_n \cdot \mathbf{S}_{n+\delta}. \tag{1}$$

Let us consider the eigenstates of this Hamiltonian when one spin deviation is present in the system. For this purpose let us introduce a complete set of orthonormal single-spin deviation states

$$|n\rangle = \begin{pmatrix} 0 \\ 0 \\ \cdot \\ \cdot \\ \cdot \\ 1 \\ \cdot \\ \cdot \\ \cdot \\ 0 \\ 0 \end{pmatrix}. \tag{2}$$

In this basis the matrix elements of $\mathcal{H}_0$ are

$$\langle n|\mathcal{H}_0|m\rangle = [-2JNS^2 + 4JS]\Delta(n,m) - 2JS\sum_{\delta=\pm 1}\Delta(n, m+\delta). \tag{3}$$

Let $|k\rangle$ be a single-spin-flip eigenstate of $\mathcal{H}_0$; then

$$\mathcal{H}_0|k\rangle = E_0(k)\,|k\rangle. \tag{4}$$

Expanding $|k\rangle$ in terms of the single-spin deviation states, and multiplying by $\langle n|$ gives a system of N homogeneous equations of the form

$$[-2JNS^2 + 4JS - E_0(k)]\langle n|k\rangle - 2JS\sum_{\delta=\pm 1}\langle n+\delta|k\rangle = 0. \tag{5}$$

Assuming periodic boundary conditions these equations have the solution

$$\langle n|k\rangle = (1/\sqrt{N})\exp(ikna), \tag{6}$$

where a is the distance between spins and k is equal to some integer times $2\pi/Na$. It is customary to choose the N values from $-(\pi/Na)(N-2)$ up to $+\pi/a$. The corresponding eigenvalue is

$$\begin{aligned} E_0(k) &= -2JNS^2 + 4JS(1-\cos ka) \\ &= E_0(\pi/2a) - 4JS\cos(ka), \end{aligned} \tag{7}$$

which is the familiar spin-wave dispersion relation.

Now let us replace the spin at l by an impurity spin S', which has an exchange coupling J' with its host neighbors. The Hamiltonian then becomes

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1, \tag{8}$$

where

$$\mathcal{H}_1 = 2(J\mathbf{S}_l - J'\mathbf{S}_l')\cdot\sum_{\delta=\pm 1}\mathbf{S}_{l+\delta}. \tag{9}$$

The matrix elements of $\mathcal{H}_1$ in the basis of single-spin deviation states are

$$\begin{aligned} \langle n|\mathcal{H}_1|m\rangle &= -4JS^2\rho\Delta(n,m) + 4JS\epsilon\Delta(l,m)\Delta(n,m) \\ &\quad + 2JS\rho\sum_\delta \Delta(l+\delta, m)\Delta(n,m) - 2JS\gamma \\ &\quad \times \sum_\delta [\Delta(l+\delta, n)\Delta(l,m) + \Delta(l,n)\Delta(l+\delta,m)], \end{aligned} \tag{10}$$

where

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

and

$$\gamma = (J'/J)(S'/S)^{1/2} - 1. \tag{11}$$

Let us redefine $\mathcal{H}_0$ and $\mathcal{H}_1$ by incorporating the first term in $\langle n|\mathcal{H}_1|m\rangle$ above into $\langle n|\mathcal{H}_0|m\rangle$. This now means that $E_0(\pi/2a) = -2NJS^2 + 4JS - 4S(J'S' - JS)$. Then $\langle n|\mathcal{H}_1|m\rangle$ is an N by N matrix, all the elements of which are zero except for a 3 by 3 submatrix centered at l. Since all the sites are equivalent we may choose $l = 2$. Then $\langle n|\mathcal{H}_1|m\rangle$ has the form

$$\langle n|\mathcal{H}_1|m\rangle = 2JS\left(\begin{array}{ccc|c} \rho & -\gamma & 0 & \\ -\gamma & 2\epsilon & -\gamma & 0 \\ 0 & -\gamma & \rho & \\ \hline & 0 & & 0 \end{array}\right). \tag{12}$$

Let $|\Psi\rangle$ be the eigenstate of the system in the presence of the impurity. This may be expanded in single-spin deviation states,

$$|\Psi\rangle = \sum_n \langle n|\Psi\rangle\,|n\rangle = \begin{pmatrix} \langle 1|\Psi\rangle \\ \langle 2|\Psi\rangle \\ \vdots \\ \langle N|\Psi\rangle \end{pmatrix}. \tag{13}$$

Thus the Schrödinger equation is the matrix equation

$$(\mathcal{H}_0+\mathcal{H}_1)\,|\,\Psi\rangle=E\,|\,\Psi\rangle, \tag{14}$$

or

$$[I-(E-\mathcal{H}_0)^{-1}\mathcal{H}_1]\,|\,\Psi\rangle=0, \tag{15}$$

where I is the N by N unit matrix. We shall find it convenient to introduce the Green's-function matrix

$$G=2JS(E-\mathcal{H}_0)^{-1}. \tag{16}$$

The eigenvalue equation then becomes

$$[I-(2JS)^{-1}G\mathcal{H}_1]\,|\,\Psi\rangle=0. \tag{17}$$

Because of the form of $\mathcal{H}_1$ the matrix product $G\mathcal{H}_1$ has elements only in its first three columns, in particular

$$1-(2JS)^{-1}G\mathcal{H}_1=\left(\begin{array}{ccc|cccccc}
1-\rho G_{11}+\gamma G_{12} & \gamma G_{11}-2\epsilon G_{12}+\gamma G_{13} & \gamma G_{12}-\rho G_{13} & & & & & & \\
-\rho G_{21}+\gamma G_{22} & 1+\gamma G_{21}-2\epsilon G_{22}+\gamma G_{23} & \gamma G_{22}-\rho G_{23} & & & & 0 & & \\
-\rho G_{31}+\gamma G_{32} & \gamma G_{31}-2\epsilon G_{32}+\gamma G_{33} & 1+\gamma G_{32}-\rho G_{33} & & & & & & \\
\hline
 & \cdot & & 1 & & & & & \\
 & \cdot & & & 1 & & & & \\
 & \cdot & & & & 1 & & & \\
-\rho G_{n1}+\gamma G_{n2} & \gamma G_{n1}-2\epsilon G_{n2}+\gamma G_{n3} & \gamma G_{n2}-\rho G_{n3} & & & & \cdot & & \\
 & \cdot & & & & & & \cdot & \\
 & \cdot & & & & & & & \cdot \\
 & \cdot & & & & & & &
\end{array}\right). \tag{18}$$

This means that the first three components of $|\,\Psi\rangle$, corresponding to the impurity and its nearest neighbors, may be solved for independent of the others. These other components are then easily found from the remaining equations. Thus our problem reduces to the 3×3 matrix equation

$$[I-(2JS)^{-1}G\mathcal{H}_1]\,|\,\Psi\rangle=0, \tag{19}$$

where I, G, and $\mathcal{H}_1$ are now 3×3 matrices and $|\,\psi\rangle$ is constructed from the first three components of $|\,\Psi\rangle$.

The matrix elements of G are

$$G_{nm}=2JS\langle n\,|\,1/E-\mathcal{H}_0\,|\,m\rangle. \tag{20}$$

Expanding the spin-deviation states in terms of the eigenfunctions of $\mathcal{H}_0$ the matrix elements become

$$G_{nm}=\frac{2JS}{N}\sum_k\frac{\exp[ik(n-m)a]}{E-E_0(k)}. \tag{21}$$

Notice that G_{nm} depends only upon $|\,n-m\,|$. Thus there are only three different matrix elements of G in this part of our problem,

$$G=\begin{pmatrix} G_0 & G_1 & G_2\\ G_1 & G_0 & G_1\\ G_2 & G_1 & G_0\end{pmatrix}. \tag{22}$$

The general matrix element G_n may be evaluated by converting the sum over k into an integral,

$$G_n=\frac{2JS}{N}\frac{Na}{2\pi}\int_{-\pi/a}^{\pi/a}\frac{\exp(ikna)\,dk}{E-E_0(\pi/2a)+4JS\cos(ka)+i4JS\epsilon}. \tag{23}$$

Introducing

$$\mathcal{E}=\frac{E-E_0(\pi/2a)}{4JS}, \tag{24}$$

this integral may be evaluated by the method of residues and gives

$$G_n(\mathcal{E})=\frac{[(\mathcal{E}^2-1)^{1/2}-\mathcal{E}]^n}{2(\mathcal{E}^2-1)^{1/2}}. \tag{25}$$

Notice that for $\mathcal{E}>1$ the Green's function is real, while it becomes complex when $\mathcal{E}<1$.

As in all problems involving clusters of ions it is convenient to work on a basis which is characterized by an irreducible representation of the point group. This is equivalent to transforming G and $\mathcal{H}$ by a unitary transformation which diagonalizes some symmetry operator of the point group. For example, the interchange of particles 1 and 3 is a symmetry operation of the system and has the representation

$$\begin{pmatrix} 0 & 0 & 1\\ 0 & 1 & 0\\ 1 & 0 & 1\end{pmatrix}. \tag{26}$$

A unitary transformation which diagonalizes this matrix is

$$U=\begin{pmatrix} 0 & 1 & 0\\ \tfrac{1}{2}\sqrt{2} & 0 & \tfrac{1}{2}\sqrt{2}\\ \tfrac{1}{2}\sqrt{2} & 0 & -\tfrac{1}{2}\sqrt{2}\end{pmatrix}. \tag{27}$$

The new states are

$$U\,|\,\psi\rangle=\begin{pmatrix} \langle 2\,|\,\Psi\rangle\\ \tfrac{1}{2}\sqrt{2}(\langle 1\,|\,\Psi\rangle+\langle 3\,|\,\Psi\rangle)\\ \tfrac{1}{2}\sqrt{2}(\langle 1\,|\,\Psi\rangle-\langle 3\,|\,\Psi\rangle)\end{pmatrix}. \tag{28}$$

Because of their symmetrical form, the first two new modes are referred to as being s-like modes while the third is a p-like mode.

The transformed Green's function and Hamiltonian lead to the determinantal equation

$$\begin{vmatrix} 1-2\epsilon G_0+2\gamma G_1 & \sqrt{2}(\gamma G_0-\rho G_1) & 0 \\ \sqrt{2}(\gamma G_0-2\epsilon G_1+\gamma G_2) & 1+2\gamma G_1-\rho(G_0+G_2) & 0 \\ 0 & 0 & 1-\rho(G_0-G_2) \end{vmatrix}=D_sD_p=0. \tag{29}$$

As we see, the p-like mode is relatively simple, depending only upon the parameter ρ. As a result of this simplicity we shall find that we can obtain analytic expressions for the eigenvalues and eigenfunctions of this mode. For this reason we shall specialize our discussion to this mode, arguing later that the results obtained apply generally to the other modes as well.

The eigenvalue is given by

$$D_p=1-\rho(G_0-G_2)=0. \tag{30}$$

Using the results for G_0 and G_2 obtained from the general relation for G_n, this eigenvalue for $\mathcal{E}>1$ becomes

$$1-[\rho/2(\mathcal{E}^2-1)^{1/2}][1-((\mathcal{E}^2-1)^{1/2}-\mathcal{E})^2]=0, \tag{31}$$

which leads to

$$\mathcal{E}=(\rho^2+1)/2\rho. \tag{32}$$

The behavior of this p-like mode is shown in Fig. 2. As ρ becomes large, the eigenvalue of this mode becomes $\rho/2$. Thus, its energy is

$$\begin{aligned} E&=E_0(\pi/2a)+2JS\rho \\ &=[-2NJS^2-4S(J'S'-JS)]+4JS+2(J'S'-JS). \end{aligned} \tag{33}$$

The part in the square bracket is just the ground-state energy of the impurity system while the remaining part is the Ising energy required to create a spin deviation on a host spin which is a neighbor of the impurity. Thus, as the local modes move away from the spin-wave band, their frequencies approach an Ising limit.

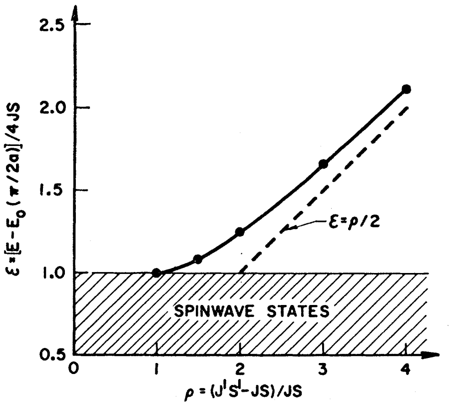

FIG. 2. Frequency of p-like local mode in a linear chain.

The eigenfunction of this p-like mode may be obtained exactly. For this mode we have

$$G_n=\frac{\rho(-1/\rho)^n}{\rho^2-1}. \tag{34}$$

From the first three of our N homogeneous equations we obtain

$$\langle 2\mid\Psi\rangle=0$$

and

$$\langle 3\mid\Psi\rangle=-\langle 1\mid\Psi\rangle.$$

Therefore, the nth equation gives for $n>3$

$$\langle n\mid\Psi\rangle=\rho(G_{n-1}-G_{n-3})\langle 1\mid\Psi\rangle=\rho^3(-1/\rho)^n\langle 1\mid\Psi\rangle. \tag{35}$$

Finally, $\langle 1\mid\Psi\rangle$ is obtained by the condition that the total spin deviation be one, i.e., that the function be normalized. This condition gives

$$\begin{aligned} &2\,|\langle 1\mid\Psi\rangle|^2+2\sum_{n=4}^{N/2}|\langle n\mid\Psi\rangle|^2 \\ &\qquad=2[1+\rho^6\sum_{n=4}^{N/2}(-1/\rho)^{2n}]\,|\langle 1\mid\Psi\rangle|^2=1, \end{aligned} \tag{36}$$

and therefore

$$|\langle 1\mid\Psi\rangle|=\left\{\frac{\rho^2-1}{2[\rho^2-1/(\rho N-6)]}\right\}^{1/2}. \tag{37}$$

As ρ goes to zero this approaches $1/\sqrt{N}$. In the limit of large N it becomes

$$|\langle 1\mid\Psi\rangle|=(1/\rho)[\tfrac{1}{2}(\rho^2-1)]^{1/2}.$$

Choosing $\langle 1\mid\Psi\rangle$ as positive we have

$$\langle 3\mid\Psi\rangle=-(1/\rho)[\tfrac{1}{2}(\rho^2-1)]^{1/2} \tag{38}$$

and

$$\langle n\mid\Psi\rangle=[\tfrac{1}{2}(\rho^2-1)]^{1/2}(-1/\rho)^{n-2},\qquad n>3.$$

The resulting wave functions are shown in Fig. 3. The origin of the term "p-like" mode is now evident. Since $|\langle n\mid\Psi\rangle|^2$ is the probability of finding the spin deviation at site n we see that as the mode moves away from the spin-wave band it becomes more localized.

Notice that the eigenvalue (32) is defined for all values of ρ. However, for $\rho<1$ the solution does not correspond to a localized mode. To see this let us consider the eigenvalue condition in its original form in-

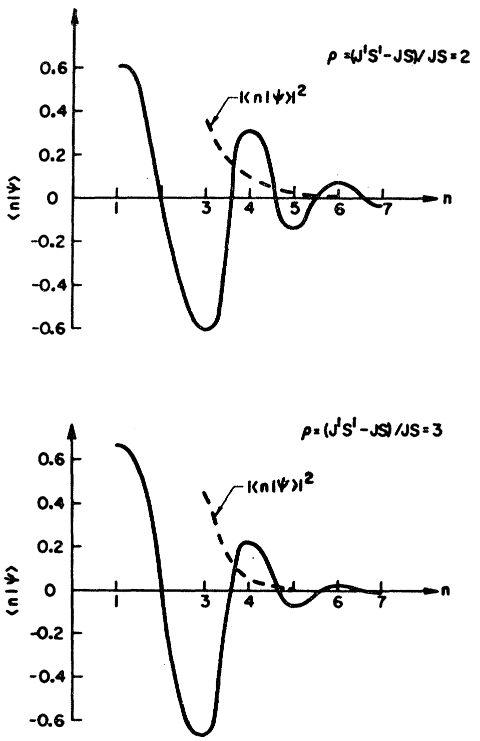

FIG. 3. Wave functions of p-like mode in a linear chain.

volving sums over k. This becomes

$$\sum_{k=2\pi/Na,4\pi/Na,\cdots}^{\pi/a} \frac{1-\cos(2ka)}{\mathcal{E}+\cos(ka)}=\frac{N}{\rho}. \tag{39}$$

The left-hand and right-hand sides of this equation are plotted schematically in Fig. 4. The solutions correspond to the intersections of these two functions. We see that when ρ is large there is always a solution at point A with $\mathcal{E}>1$. As ρ decreases the horizontal line N/ρ moves upward, eventually intersecting at $\mathcal{E}=1$ (point B). The value of ρ at which this occurs is obtained by evaluating the sum on the left-hand side of Eq. (39) with $\mathcal{E}=1$. This may be accomplished by grouping the terms for $k=n2\pi/Na$ with those for $k=(N/2-n)2\pi/Na$. The result is that the sum has the value N. Thus we find that when $\rho=1$ the solution just begins to enter what was the original unperturbed spin-wave band. At this point the separation of this solution from the next solution at B' becomes comparable to the separation between the spin-wave states themselves. Therefore, any formalism employing an integral representation of the Green's functions is not capable of distinguishing local modes from band modes beyond this point. In order to describe the situation for $\rho<1$ we must consider the density of states. The density of states $\eta(\mathcal{E})$ of the perturbed system is related to the Green's function $\mathcal{G}(\mathcal{E})$ of the perturbed system by

$$\eta(\mathcal{E})=-(2/\pi)\ \mathrm{Im}\mathrm{Tr}\mathcal{G}(\mathcal{E}+i\epsilon). \tag{40}$$

The perturbed Green's function is defined analogously to the unperturbed Green's function, i.e.,

$$\mathcal{G}=2JS(E-\mathcal{H})^{-1}. \tag{41}$$

This is related to G by the identity

$$\begin{aligned}\mathcal{G}&=[1-(2JS)^{-1}G\mathcal{H}_1]^{-1}G\\ &=G+(2JS)^{-1}G\mathcal{H}_1[1-(2JS)^{-1}G\mathcal{H}_1]^{-1}G.\end{aligned} \tag{42}$$

Taking the trace, we have

$$\begin{aligned}\mathrm{Tr}\mathcal{G}=\mathrm{Tr}\mathrm{G}+\sum_{nlmp} G_{nl}(2JS)^{-1}(\mathcal{H}_1)_{lm}\\ \times[1-(2JS)^{-1}G\mathcal{H}_1]_{mp}{}^{-1}G_{pn}.\end{aligned} \tag{43}$$

This may be written as[8]

$$\mathrm{Tr}\mathcal{G}=\mathrm{Tr}G+(1/2D)(d/d\mathcal{E})D, \tag{44}$$

where

$$D=|1-(2JS)^{-1}G\mathcal{H}_1|. \tag{45}$$

Therefore, we have

$$\eta(\mathcal{E})=\eta_0(\mathcal{E})-(1/\pi)\ \mathrm{Im}(d/d\mathcal{E})\ \ln D, \tag{46}$$

where the unperturbed density of states is given by

$$\begin{aligned}\eta_0(\mathcal{E})&=-(2/\pi)\ \mathrm{Im}\mathrm{Tr}G(\mathcal{E}+i\epsilon)\\ &=-(2/\pi)\ \mathrm{Im}NG_0(\mathcal{E}+i\epsilon)\\ &=N/\pi(1-\mathcal{E}^2)^{1/2}.\end{aligned} \tag{47}$$

The singularities at $\mathcal{E}=\pm1$ are due, of course, to the one-dimensional nature of our problem. The contribution of the p mode to the density of states may be determined explicitly by recognizing that the deter-

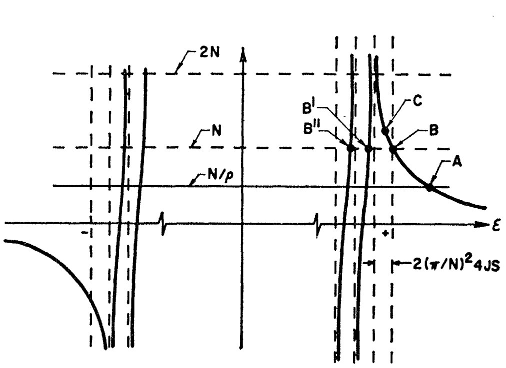

FIG. 4. Schematic description of local-mode solution.

---

[8] Y. Isyumov, Advan. Phys. 14, 569 (1965).

minant D factors into an s part and a p part, thus

$$\eta_p=-(1/\pi)\,\mathrm{Im}(D_p'/D_p)$$
$$=\frac{\mathrm{Im}D_p\,\mathrm{Re}D_p'-\mathrm{Re}D_p\,\mathrm{Im}D_p'}{\pi[(\mathrm{Re}D_p)^2+(\mathrm{Im}D_p)^2]}. \tag{48}$$

For $\mathcal{E}<1$, D_p becomes

$$D_p=1-\rho\mathcal{E}+i\rho(1-\mathcal{E}^2)^{1/2}, \tag{49}$$

and therefore

$$\eta_p=\rho(\mathcal{E}-\rho)/\pi(1-\mathcal{E}^2)^{1/2}(1-2\rho\mathcal{E}+\rho^2). \tag{50}$$

This is plotted in Fig. 5 as a function of $\mathcal{E}$ for $\rho=4$ and $\rho=\frac{1}{2}$. The unperturbed density of states divided by N is indicated by the dashed curve. Notice that

$$\begin{aligned}\int_{-1}^{+1}\eta_p(\mathcal{E})\,d\mathcal{E}&=0 \qquad \rho<1\\ &=-1 \qquad \rho>1.\end{aligned} \tag{51}$$

This minus one corresponds to the fact that the localized mode appearing outside the band does so at the expense of one mode inside the band. We see that the construction of the local mode results in a nearly uniform depletion of states within the band. For $\rho<1$ the perturbation is not strong enough to produce either a localized mode or a resonant state, but merely causes a redistribution of the states.

In the three-dimensional case when a mode appears inside the band it manifests itself as a definite "bump" in the density of states. Presumably, the one-dimensional nature of our problem is responsible for the absence of such a bump in this case.

III. EIGENVALUES FOR TWO IMPURITIES

Consider, in addition to the first impurity at site 2, a second identical impurity at the site $n+2\geq 5$. Thus $n-1$ is the number of intervening host spins. The perturbed Hamiltonian then becomes

$$\mathcal{H}_1=2JS\begin{array}{c}\begin{matrix}1 & 2 & 3 & \cdots & n+1 & n+2 & n+3\end{matrix}\\ \begin{pmatrix}\rho & -\gamma & 0 & & & & \\ -\gamma & 2\epsilon & -\gamma & & & & \\ 0 & -\gamma & \rho & & & & \\ & & & \ddots & & & \\ & & & & \rho & -\gamma & 0\\ & & & & -\gamma & 2\epsilon & -\gamma\\ & & & & 0 & -\gamma & \rho\end{pmatrix}\end{array}. \tag{52}$$

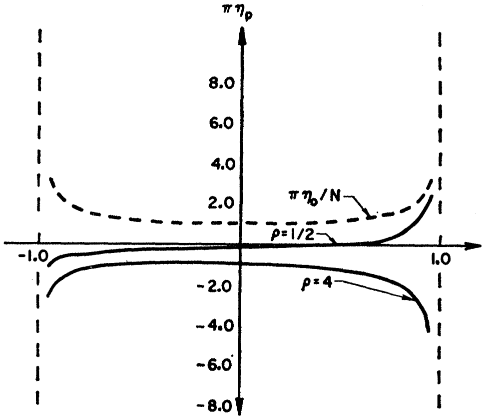

FIG. 5. In-band density of states for p-like mode.

The reduced eigenvalue equation in this case becomes

$$[I-(2JS)^{-1}G\mathcal{H}_1]\,|\psi\rangle=0, \tag{53}$$

where

$$G=\begin{pmatrix}G_0 & G_1 & G_2 & G_n & G_{n+1} & G_{n+2}\\ G_1 & G_0 & G_1 & G_{n-1} & G_n & G_{n+1}\\ G_2 & G_1 & G_0 & G_{n-2} & G_{n-1} & G_n\\ G_n & G_{n-1} & G_{n-2} & G_0 & G_1 & G_2\\ G_{n+1} & G_n & G_{n-1} & G_1 & G_0 & G_1\\ G_{n+2} & G_{n+1} & G_n & G_2 & G_1 & G_0\end{pmatrix}, \tag{54}$$

and

$$\mathcal{H}_1=2JS\left(\begin{array}{ccc:ccc}\rho & -\gamma & 0 & & & \\ -\gamma & 2\epsilon & -\gamma & & 0 & \\ 0 & -\gamma & \rho & & & \\ \hdashline & & & \rho & -\gamma & 0\\ & 0 & & -\gamma & 2\epsilon & -\gamma\\ & & & 0 & -\gamma & \rho\end{array}\right)$$

and

$$|\psi\rangle=\begin{pmatrix}\langle 1\,|\,\Psi\rangle\\ \langle 2\,|\,\Psi\rangle\\ \langle 3\,|\,\Psi\rangle\\ \langle n-1\,|\,\Psi\rangle\\ \langle n\,|\,\Psi\rangle\\ \langle n+1\,|\,\Psi\rangle\end{pmatrix}. \tag{55}$$

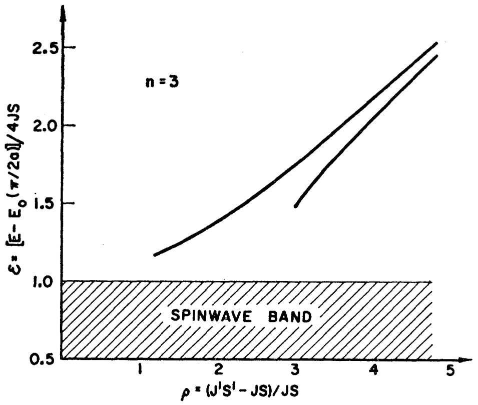

FIG. 6. Splitting of p-like mode with two impurities.

Just as in the case of one impurity we can block-diagonalize the eigenvalue matrix by making use of symmetry operations. Again, the s-like modes are extremely complicated while the p-like modes are determined by

$$D_p^{(+)}D_p^{(-)}=0, \tag{56}$$

where

$$D_p^{(\pm)}=1-\rho\{G_0-G_2\pm[G_n+\tfrac{1}{2}(G_{n+2}+G_{n-2})]\}. \tag{57}$$

For $\mathcal{E}>1$ this gives

$$\rho=\frac{2(\mathcal{E}^2-1)^{1/2}}{1-[(\mathcal{E}^2-1)^{1/2}-\mathcal{E}]^2\pm 2\mathcal{E}^2[(\mathcal{E}^2-1)^{1/2}-\mathcal{E}]^n}. \tag{58}$$

For a given value of ρ there are two solutions, shown in Fig. 6 for $n=3$. The splitting of these two solutions, $\Delta\mathcal{E}$, is shown in Fig. 7 as a function of n. By comparing Fig. 7 with Fig. 4 we can see that the coupling between two localized modes appears to depend upon the overlap of their wave functions.

If the perturbation for $\rho<1$ were strong enough to produce an identifiable mode within the band, i.e., a bump, then we could proceed in the same way to find the splitting of such a mode and thereby determine the interaction between in-band modes. However, the one-dimensional nature of our problem seems to preclude this possibility.

IV. PERTURBATION THEORY

It is interesting to compare results with those obtained from a spin-wave scattering approach. Consider impurities from the single-ion point of view. The longitudinal part of the exchange interaction between an impurity and the host produces a molecular field, while the transverse part enables the impurity to emit or absorb spin waves.

For our one-dimensional chain of N spins, the Hamiltonian is given by

$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}_1, \tag{59}$$

where

$$\mathcal{H}_0=-2JNS^2+\sum_k \epsilon_k(a_k^\dagger a_k+\tfrac{1}{2})-4J'\langle S^z\rangle S_2'^z -4J'\langle S^z\rangle S_{n+2}'^z \tag{60}$$

and

$$\mathcal{H}_1=-2(2S/N)^{1/2}J'S_2^\dagger \sum_k \gamma_k \exp(-ik2a)a_k^\dagger +\text{c.c.}-2(2S/N)^{1/2}J'S_{n+2}'^\dagger \sum_k \gamma_k \exp[-ik(n+2)a]a_k^\dagger +\text{c.c.} \tag{61}$$

Here a_k and $a_k^\dagger$ are the spin-wave annihilation and creation operators, respectively, and are related to the spins $\mathbf{S}_n$ in the usual manner. The spin-wave energy is $\epsilon_k=4JS[1-\cos(ka)]$ and

$$\gamma_k=\tfrac{1}{2}\sum_{\delta=\pm1}\exp(ik\delta a).$$

We shall take the thermal average $\langle S^z\rangle$ to be S. Notice that we are neglecting any effect which the impurity may have on the original spin-wave spectrum.

We now apply second-order perturbation theory to obtain an effective interaction between the impurity spins. Those terms leading to coupling between the spins are

$$-2J_{\text{eff}}^{(n)}(S_2'^-S_{n+2}'^+ + S_2'^+S_{n+2}'^-), \tag{62}$$

where

$$J_{\text{eff}}^{(n)}=\frac{4S}{N}J'^2\sum_k\frac{\cos^2(ka)\exp(ikna)}{4J'S+\epsilon_k}. \tag{63}$$

This anisotropic exchange interaction leads to a split-

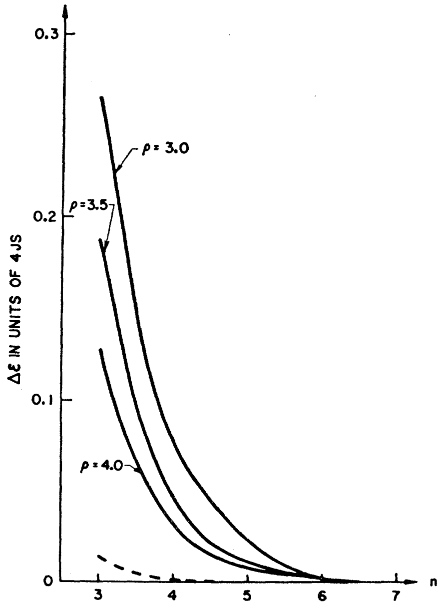

FIG. 7. Mode splitting as a function of impurity separation.

ting of the single-ion levels equal to $J_{\text{eff}}(n)$. This result differs from the Suhl-Nakamura result in two respects. First of all, since we are dealing with electronic spins instead of nuclear spins we must retain the impurity spin-flip energy in the denominator. Secondly, there is the $\cos^2(ka)$ factor which arises from the fact that the impurity spin couples to the host via a nearest-neighbor exchange interaction instead of a contact interaction.

Converting the sum in $J_{\text{eff}}^{(n)}$ into an integral, the mode splitting is given in units of $4JS$ as

$$\Delta\mathcal{E}=\frac{1}{4S}\,\frac{(1+\epsilon)^2(3+\epsilon)\{2+\epsilon-[(2+\epsilon)^2-1]^{1/2}\}^n}{[(2+\epsilon)^2-1]^{1/2}},\qquad(64)$$

where ϵ is defined as

$$\epsilon\equiv(J'-J)/J=(1+\rho)(S/S')-1.$$

This result is plotted as the dashed curve in Fig. 7 as a function of n for $S'=S=\frac{1}{2}$ and $\rho=3$, 3.5, and 4.

We see that the result obtained from a spin-wave scattering calculation differs appreciably from the exact solution. Part of this difference undoubtedly arises from the fact that our exact results apply to a p-like localized mode whereas an s-like mode calculation provides a more appropriate comparison with the scattering theory. Nevertheless, we feel that our exact determination of the impurity coupling illustrates that *bound states*, which scattering theory neglects, are very important.

V. SIMPLE CUBIC LATTICE

An impurity spin in a simple cubic Heisenberg ferromagnet gives rise to a 7×7 perturbation matrix. This factors into two s-like modes, three p-like modes, and two d-like modes. An analytic representation for the eigenvalues of any of these modes analogous to Eq. (32) is not possible since the three-dimensional Green's function is an integral over three Bessel functions. The numerical results for the triply degenerate p-like mode are shown in Fig. 8. Again we notice that as the mode moves away from the spin-wave band, its frequency approaches the Ising result for a spin deviation at a site adjacent to the impurity.

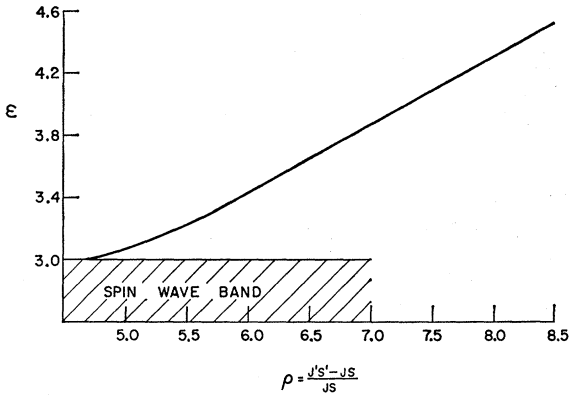

FIG. 8. Frequency of p-like local mode in a simple cubic lattice.

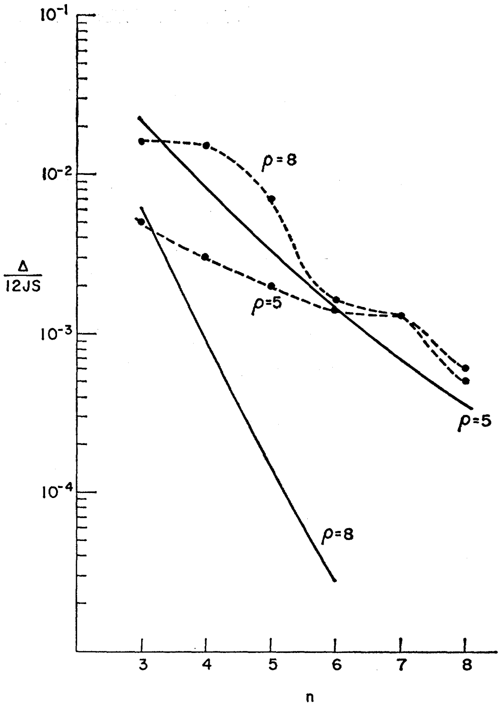

FIG. 9. The p-like mode splitting for the simple cubic lattice.

If we now add a second impurity separated by n lattice spacings along, say, the x axis, then the p_x mode splits. This splitting as a function of n is shown by the solid curves in Fig. 9. We see that the mode splitting decreases very rapidly as the mode moves away from the spin-wave band. The dashed curves in Fig. 9 are the results of a perturbation calculation analogous to the one-dimensional calculation described in Sec. IV. Again we see that the neglect of bound states gives results which differ appreciably from the exact results as the modes move away from the band.