PHYSICAL REVIEW B VOLUME 27, NUMBER 5 1 MARCH 1983

Theory of the response of local magnon modes in antiferromagnets

Sergio M. Rezende
Departamento de Física, Universidade Federal de Pernambuco, 50.000 Recife, Brasil

(Received 16 March 1982; revised manuscript received 10 February 1983)

The features of the local magnon modes associated with magnetic impurities in antiferromagnetic insulators are studied in detail. A Green's-function theory is used to calculate the frequency, the wave functions, and the dynamic response at $T=0$ in the limit of very small impurity concentrations and at arbitrary values of the applied magnetic field. The theory is applied to MnF_2:Fe^{2+} and to FeF_2:Mn^{2+} and it explains quantitatively the large enhancement of Mn impurity modes in the gap of antiferromagnetic FeF_2. In this system the local mode intensity is strongly dependent on the competition between the nearest neighbor J_1' and the intersublattice J_2' Mn-Fe interaction parameters. This allows one to determine the interaction parameters from Raman scattering data. Application to FeF_2:Mn yields $|J_1'|=0.2$ cm^{-1} and $J_2'=1.79$ cm^{-1}, both antiferromagnetic, and this confirms previous evidence that the two interactions compete in this system.

I. INTRODUCTION

The excitations in randomly disordered magnetic systems have been the subject of continuing interest for many years. Initial attention was focused on the local modes associated with the defects in systems with low impurity concentrations. The antiferromagnetic fluorides received particular attention and were investigated by a number of experimental techniques, such as neutron scattering,[1,2] Raman light scattering,[3,4] far-infrared spectroscopy,[5] optical absorption and fluorescence,[6] and nuclear magnetic resonance.[7] This early work on the fluorides was mainly directed to systems in which the localized modes have energies above the host spin-wave band, and was reviewed in detail by Cowley and Buyers.[8] Relatively little work was done on systems in which a local mode lies in the antiferromagnetic gap, such as CoF_2:Mn and FeF_2:Mn. The former system was investigated by neutron[9,10] and light scattering,[11] but apparently not much attention was paid to the anomalously high intensity of the gap modes in the earlier studies.

With the development of far-infrared lasers, high-resolution studies of impurity modes became possible and a number of antiferromagnetic systems have recently been investigated in greater detail.[12–15] The Mn impurity modes in CoF_2 and in FeF_2 have been found to be greatly enhanced due to their proximity to the edge of the spin-wave band ($k=0$ magnon). The frequencies of the impurity and $k=0$ host modes, as well as their linewidths and intensities have been studied in great detail[15] in FeF_2:Mn. This system has also been studied by Raman light scattering[16] that shows an enhancement of the impurity mode which is even more dramatic. This is so because one-magnon light scattering is usually not observed in manganese compounds due to the weak interaction between the Mn^{2+} spins and the radiation. But when minute amounts of Mn^{2+} are substitutionally introduced in FeF_2 the impurity mode becomes as intense as the host mode of the Fe^{2+} spins. This effect resembles the anomalously high intensity observed in the light scattered from certain molecules adsorbed onto metal surfaces.[17]

The experiments in FeF_2:Mn have been explained[15] by a coupled equations-of-motion mean-field calculation that accounts satisfactorily for the influence of magnetic polaritons in the absorption line shapes, the host-impurity frequency pulling, and the enhancement of the impurity mode. In this approach one assigns different magnetizations to the impurity and to the host spins, and the normal mode frequencies are the same as those given by the Ising model. This model is known to fail when the impurity mode frequency is close to the magnon band, so one does not expect that the interaction constants determined by the calculation are the true ones.

In this paper we present a theory for the enhancement of the impurity associated local mode, which takes into account the spatial extended character of the mode. The theory uses zero-temperature Green's functions and allows for the existence of two interaction constants between the impurity and host spins, since they have been found necessary to explain several results in the (FeMn)F_2 system.

In Sec. II we formulate the spin-wave problem for an antiferromagnet with a single substituted impurity spin in order to obtain the eigenfrequencies and eigenfunctions of the impurity associated local modes. Section III is devoted to the calculation of the dynamic response of the local modes. In Sec. IV we apply the results to the systems FeF_2:Mn and MnF_2:Fe. In the last section we present the concluding remarks. Preliminary results for FeF_2:Mn have been previously presented.[18]

II. IMPURITY MODES IN ANTIFERROMAGNETS

We write the Hamiltonian for FeF_2 with a single Mn^{2+} impurity at site $i=0$, or for MnF_2:Fe^{2+}, as

$$\begin{aligned}\mathscr{H}=&\mu_B H_0 \sum g_l S_l^z + 2J\sum \vec{S}_i\cdot\vec{S}_{i+\delta}\\ &-D\sum (S_l^z)^2 + 2J_2'\vec{S}_0\cdot\sum \vec{S}_{\delta_2}\\ &-2J_1'\vec{S}_0\cdot\sum\vec{S}_{\delta_1} - D'(S_0^z)^2\ ,\end{aligned}\tag{1}$$

where the sums in l run over sublattices i and j but exclude $i=0$. The first term is the Zeeman interaction due to an external field H_0 applied along the c axis of the crystal (z direction). The second represents the Heisenberg exchange interaction between host spins which are next-nearest neighbors (on opposite spin sublattices) in the rutile structure. As is well known, the interaction J_1 between nearest neighbors in pure FeF_2 and MnF_2 is very small. The third term is the anisotropy interaction which is large and mainly of single-ion origin for Fe^{2+} and small and of dipolar origin for Mn^{2+}. Since the dipolar interaction is very small compared to exchange, we write it in the form (1) for simplicity. The last three terms describe the impurity spin interactions. The impurity spin is assumed to interact with its eight next-nearest host neighbors δ_2 (interaction parameter J_2') and with its two nearest neighbors δ_1 (parameter J_1') since there is experimental evidence that both interactions are important in the impure system. Note that the interaction parameter J_1' is taken positive when ferromagnetic.

The frequencies of local modes associated with impurities in antiferromagnets were first calculated by Lovesey[19] and Tonegawa.[20] Tonegawa[21] and Shiles and Hone[22] later extended the calculations for impurities with two interaction constants, such as the one considered here. We present here the calculation of the frequencies not only for completeness but also because we need to obtain the wave functions to find the dynamic response. To our knowledge these have not been calculated explicitly for actual systems of interest.

We assume that the dominant interaction on the impurity spin S' at site $i=0$ is the antiferromagnetic J_2' (>0), so that in the ground state it points in the positive z direction, while its eight δ_2 neighbors are in the down sublattice. The Hamiltonian (1) can be diagonalized by first introducing spin deviation operators a_i and b_j associated with the two sublattices and then transforming them into normal mode boson operators by means of

$$\begin{aligned}c_\alpha&=\sum_i \Gamma_i^\alpha a_i + \sum_j \Gamma_j^\alpha b_j^\dagger\ ,\\ c_\beta^\dagger&=\sum_i \Gamma_i^\beta a_i + \sum_j \Gamma_j^\beta b_j^\dagger\ .\end{aligned}\tag{2}$$

The wave functions obey the orthonormality relations

$$\sum_i \Gamma_i^\lambda \Gamma_i^{\lambda'*} - \sum_j \Gamma_j^\lambda \Gamma_j^{\lambda'*} = \pm\delta_{\lambda\lambda'}\ ,\tag{3}$$

where the positive sign holds for the α mode and the negative for the β one. The equations of motion for the wave functions which diagonalize the Hamiltonian can be written as

$$\underline{\Gamma} - \underline{G}^0\underline{V}\underline{\Gamma} = 0\ ,\tag{4}$$

where $\underline{\Gamma}$ is a $2N\times 1$ column matrix,

$$\underline{\Gamma} = \begin{bmatrix}\underline{\Gamma}_i\\ \underline{\Gamma}_j\end{bmatrix}\ .\tag{5}$$

$\underline{G}^0=(\underline{\omega}-\underline{H})^{-1}$ is the pure-crystal Green's-function matrix, $\underline{H}$ is the pure-crystal Hamiltonian, and $\underline{V}$ is the impurity perturbing potential. This set of secular equations gives the normal mode frequencies and wave functions, subject to the condition of Eq. (3). Owing to the finite range of the perturbation caused by the impurity the solution of Eq. (4) reduces to solving a set of 11×11 secular equations. The calculation of the corresponding determinant can be simplified by block diagonalization of the matrix $\underline{G}^0\underline{V}$. This can be accomplished via a unitary transformation from a basis of individual sites to linear combinations that transform according to the irreducible representations of the group of the cluster formed by the impurity and its interacting neighbors. The 11×11 unitary transformation for this case has been given by Shiles and Hone.[22] Instead of the site wave functions a more convenient set of normal mode amplitudes[21] can be used to solve (4). They are represented by the 11×11

column matrix $\underline{A}$ obtained from $\underline{\Gamma}$ by the transformation,

$$\underline{A}=\underline{V}\underline{\Gamma}\ . \tag{6}$$

Equations (4) and (6) lead to a new set of secular equations,

$$(\underline{I}-\underline{V}\,\underline{G}^0)\underline{A}=0\ . \tag{7}$$

These determine the eigenfrequencies and the normal mode amplitudes associated with the impurity A_0 and its interacting neighbors A_{δ_1} and A_{δ_2}, subject to the orthonormality condition (3). With the knowledge of these amplitudes and the crystal Green's functions one can find the wave function at any site in the crystal. From (6) and (7) one has

$$\underline{\Gamma}=\underline{G}^0\underline{A}\ ,$$

or

$$\Gamma_l^\lambda(\pm\omega)=G_{l0}^0(\omega)A_0^\lambda+\sum_{\delta_1}G_{l\delta_1}^0(\omega)A_{\delta_1}^\lambda+\sum_{\delta_2}G_{l\delta_2}^0(\omega)A_{\delta_2}^\lambda\ . \tag{8}$$

There are four types of pure-crystal Green's functions that can be expressed in terms of their Fourier transforms,

$$G_{ml}^{0(\sigma)}(\omega)=\frac{1}{N}\sum_k e^{i\vec{k}\cdot(\vec{r}_m-\vec{r}_l)}G_k^{(\sigma)}(\omega)\ , \tag{9}$$

where $\sigma=1$ for m and l in sublattice i, $\sigma=2$ for $m=i$, $l=j$, $\sigma=3$ for $m=j$, $l=i$, and $\sigma=4$ for $m=j$, $l=j'$. These Green's functions can be easily obtained,

$$G_k^{(1)}(\omega)=\frac{\omega'+\omega_E+\omega_A}{\omega'^2-\omega_k^2}\ ,$$

$$G_k^{(2)}(\omega)=-G_k^{(3)}(\omega)=-\frac{\gamma_k\omega_E}{\omega'^2-\omega_k^2}\ , \tag{10}$$

$$G_k^{(4)}(\omega)=\frac{\omega'-(\omega_E+\omega_A)}{\omega'^2-\omega_k^2}\ ,$$

where $\omega'=\omega+\omega_H$, $\hbar\omega_H=g\mu_BH_0$, $\hbar\omega_E=2Sz_2J$, $\hbar\omega_A=(2S-1)D$, γ_k is the usual structure factor which appears in the spin-wave frequencies,

$$\omega_{\substack{\alpha\\ \beta}}=\omega_k\pm\omega_H\ ,$$

$$\omega_k=[(\omega_E+\omega_A)^2-\gamma_k^2\omega_E^2]^{1/2}\ .$$

The cluster formed by the impurity, its two nearest, and its eight next-nearest neighbors in the rutile structure belongs to the symmetry group D_{4h} whose irreducible representations are $3A_{1g}+2A_{2u}+B_{1u}+B_{2g}+E_g+E_u$. The three A_{1g} modes have s-like wave functions which are even and nonzero at the impurity site. For the s modes we make $A_{\delta_1}\equiv A_1$ and $A_{\delta_2}\equiv A_2$ and Eq. (7) leads to

$$\begin{bmatrix} Q_{11} & Q_{12} & Q_{13}\\ Q_{21} & Q_{22} & Q_{23}\\ Q_{31} & Q_{32} & Q_{33}\end{bmatrix}\begin{bmatrix}A_0\\ A_1\\ A_2\end{bmatrix}=0\ , \tag{11}$$

where

$$\begin{aligned}
Q_{11}&=1-\omega^+(\nu U_{00}-\alpha_1\sqrt{\beta}\,U_{01}/4)+\gamma_2U'_{20}\ ,\\
Q_{12}&=2\omega^+[\alpha_1\sqrt{\beta}(U_{00}+U_{11'})/8\\
&\qquad-(\nu+\omega^-\gamma_2)U_{10}]\ ,\\
Q_{13}&=8(\gamma_2\omega^-+\nu)(1-\omega^+\omega^-U_{00})\\
&\qquad+2\alpha_1\sqrt{\beta}\,\omega^+\omega^-U_{01}\ ,\\
Q_{21}&=\omega^+\beta\alpha_1(U_{00}/\sqrt{\beta}-U_{01})/8\ ,\\
Q_{22}&=1+\omega^+\beta\alpha_1(2U_{10}/\sqrt{\beta}-U_{00}-U_{11'})/8\ ,\\
Q_{23}&=-\beta\alpha_1[\omega^+\omega^-(U_{01}-U_{00}/\sqrt{\beta})+1/\sqrt{\beta}]\ ,\\
Q_{31}&=-(\gamma_2\omega^+U_{00}-\rho U'_{20})/8\ ,\\
Q_{32}&=-\omega^+(\gamma_2+\rho\omega^-)U_{10}/4\ ,\\
Q_{33}&=1+(\rho\omega^-+\gamma_2)U'_{20}\ ,
\end{aligned} \tag{12}$$

$$\begin{aligned}
U_{ii'}&=\frac{\omega_E}{\omega^+}G_{ii'}^{(1)}=\frac{1}{N}\sum_k\frac{e^{i\vec{k}\cdot\vec{r}_{ii'}}}{(\omega'^2-\omega_k^2)/\omega_E^2}\ ,\\
U'_{ji}&=-\omega_EG_{ij}^{(2)}=\frac{1}{N}\sum_k\frac{\gamma_ke^{i\vec{k}\cdot\vec{r}_{ij}}}{(\omega'^2-\omega_k^2)/\omega_E^2}\ ,
\end{aligned} \tag{13}$$

where $\beta=S'/S$, $\alpha_1=J'_1/J$, $\alpha_2=J'_2/J$, $\gamma_2=(\alpha_2\sqrt{\beta}-1)$, $\rho=\alpha_2\beta-1$, $\delta'=(2S'-1)D'/2Sz_2J$, $\delta=\omega_A/\omega_E$,

$$\nu=(g-g')\mu_BH_0/\hbar\omega_E+\delta'\beta-\delta+\alpha_2-1+\alpha_1/4\ ,$$

and $\omega^\pm=(\omega'\pm\omega_A\pm\omega_E)/\omega_E$. 0 is the impurity site, 1 and 1′ are the nearest neighbors in the up sublattice, and 2 represents the next-nearest neighbors in the down sublattice. The frequencies of the s modes are given by

$$\det(Q)=0\ . \tag{14}$$

Note that the elements of the determinant Q for $H_0=0$ are not exactly the same as the ones given by Shiles and Hone.[22] This is so because we work with spin deviation operators whereas they use the spin operators themselves. However, the zeros of the determinant are the same as they should be because the two treatments must give the same energies at

$T=0$. With (11) we can also obtain relations between the normal mode amplitudes for the s modes,

$$\begin{aligned} A_1 &= a_1 A_0 , \\ A_2 &= a_2 A_0 , \end{aligned} \tag{15}$$

where

$$a_1 = \frac{Q_{11}Q_{23}-Q_{13}Q_{21}}{Q_{22}Q_{13}-Q_{12}Q_{23}} ,$$

$$a_2 = \frac{Q_{21}Q_{12}-Q_{11}Q_{22}}{Q_{22}Q_{13}-Q_{12}Q_{23}} .$$

Using (15) and various relations between the Green's functions[20] the wave functions for the s modes can be written as

$$\begin{aligned} \Gamma_i^s &= A_0^s \frac{\omega^+}{\omega_E} \left[(1+8a_2\omega^-)U_{i0} + a_1 U_{i1} \right. \\ &\qquad \left. + a_1 U_{i1'} - \frac{8a_2}{\omega^+}\delta_{i0} \right] , \\ \Gamma_j^s &= \frac{A_0^s}{\omega_E} [(1+8a_2\omega^-)U'_{j0} \\ &\qquad + a_1 U'_{j1} + a_1 U'_{j1'}] . \end{aligned} \tag{16}$$

The amplitude A_0^s can be obtained using (16) in the normalization condition (3):

$$A_0^s = \frac{\omega_E}{P} , \tag{17}$$

where

$$\begin{aligned} P^2 = {} & (8a_2)^2 + (1+8a_2\omega^-)^2 S_{00} \\ & + 2a_1^2(S_{11}+S_{11'}) \\ & + 4(1+8a_2\omega^-)(a_1 S_{10} - 4a_2\omega^+ U_{00}) \\ & - 32a_1a_2\omega^+ U_{01} \end{aligned}$$

and

$$S_{ii'} = \frac{\omega_E^4}{N} \sum_k \frac{\omega^{+2}-\gamma_k^2}{(\omega'^2-\omega_k^2)^2} e^{i\vec{k}\cdot(\vec{r}_i-\vec{r}_{i'})} . \tag{18}$$

Numerical results for the frequencies and wave functions of the s_0 mode will be given in Sec. IV for the (FeMn)F_2 system.

III. DYNAMIC RESPONSE OF THE LOCAL s MODES

In this section we derive an expression for the intensity of the response of the local s modes in far-infrared laser absorption and in Raman light scattering experiments. In both cases the Hamiltonian for the interaction between the external excitation and the spin system can be written as

$$\mathscr{H}_1 = -\sum_{l,\alpha} m_l S_l^{\alpha\dagger} H_l^\alpha e^{-i\omega t} , \tag{19}$$

where in the far-infrared absorption $H_l^\alpha e^{-i\omega t}$ is the α component of the magnetic field of the radiation and $m_l = g_l\mu_B$. In the Raman scattering experiment H_l^α is an effective field[23] proportional to the product of the electric fields of the incident laser and of the scattered light, $\omega = |\omega_L - \omega_s|$ is the Raman shift, and m_l expresses the strength of the coupling between the radiation fields and the spin $\vec{S}_l$. The $++$ component of the susceptibility of the spin system for uniform ($q=0$) excitation is given by[24]

$$\chi^{++}(\omega, q=0) = \frac{\pi}{\hbar V} \sum_{l,l'} m_l m_{l'} \langle\langle S_l^+ ; S_{l'}^- \rangle\rangle_\omega , \tag{20}$$

where V is the volume of the crystal and the term in double angle brackets is the time Fourier transform of the retarded Zubarev[25] Green's functions for the positive and negative components of the spin operators. In order to find the susceptibility one must solve the equations of motion for the Green's functions of the impure spin system. This is easily done if the spin operators are expressed in terms of the normal mode operators c_λ and $c_\lambda^\dagger$ by means of the inverse of (2). The equations of motion[25] for the Green's functions of the various pairs of operators c_λ and $c_\lambda^\dagger$ can be solved exactly, leading to

$$\begin{aligned} \langle\langle c_\lambda ; c_{\lambda'}^\dagger \rangle\rangle_\omega &= \frac{1}{2\pi} \frac{\delta_{\lambda\lambda'}}{\omega-\omega_\lambda} , \\ \langle\langle c_\lambda^\dagger ; c_{\lambda'} \rangle\rangle_\omega &= \frac{-1}{2\pi} \frac{\delta_{\lambda\lambda'}}{\omega+\omega_\lambda} , \\ \langle\langle c_\lambda ; c_{\lambda'} \rangle\rangle_\omega &= \langle\langle c_\lambda^\dagger ; c_{\lambda'}^\dagger \rangle\rangle_\omega = 0 . \end{aligned} \tag{21}$$

The Green's functions appearing in (20) can be obtained with (2) and (21):

$$\begin{aligned} & \langle\langle S_l^+ ; S_{l'}^- \rangle\rangle_\omega \\ & \quad = \pm \frac{(S_l S_{l'})^{1/2}}{\pi} \left[\sum_\alpha \Gamma_l^{\alpha*} \Gamma_{l'}^\alpha \frac{1}{\omega-\omega_\alpha} \right. \\ & \qquad \left. - \sum_\beta \Gamma_l^{\beta*} \Gamma_{l'}^\beta \frac{1}{\omega+\omega_\beta} \right] , \end{aligned} \tag{22}$$

where the plus sign holds for l and l' in the same sublattice and the minus sign for l and l' in opposite sublattices. The result for the susceptibility depends now on the knowledge of the wave functions. For the simple case of a pure crystal, with $S_l = S$, $m_l = g\mu_B$, the susceptibility becomes

$$\chi_0^{++}(\omega, q=0) = \frac{2\gamma M \omega_A}{(\omega+\omega_H)^2 - \omega_0^2} , \tag{23}$$

where $\gamma M = Ng^2\mu_B^2 S/\hbar V$ and ω_0 is the $k=0$ magnon frequency in the absence of the field H_0. This is the well-known result[15] for the magnetic susceptibility of a uniaxial antiferromagnet under a circularly polarized rf field perpendicular to the easy axis. In the case of a crystal with an impurity at $i=0$ the wave functions can be related to the normal mode amplitudes of the impurity cluster by means of Eq. (8). Then the sums in (20) over the whole crystal can be reduced to sums of the pure-crystal Green's functions which are evaluated exactly. For a uniform excitation they will all be related to the $q=0$ components of the Green's functions (10). In order to calculate the sums in (20) we add and subtract to all terms involving the impurity site a term with the host spin parameters. We are then left with two sums, one that contains the impurity parameters and one that does not. For ω near the frequency of a λ local mode, only the terms with a resonant denominator need to be kept, and the susceptibility becomes

$$\chi^{++}(\omega \cong \omega_\lambda, q=0) = \chi_1 + \chi_2 , \tag{24}$$

where

$$\chi_1 = \frac{m^2 S}{\hbar V} \frac{1}{\omega - \omega_\lambda} \sum_{\substack{ii' \\ jj'}} (\Gamma_i^{\lambda *}\Gamma_{i'}^{\lambda} - \Gamma_i^{\lambda *}\Gamma_j^{\lambda} - \Gamma_j^{\lambda *}\Gamma_i^{\lambda} + \Gamma_j^{\lambda *}\Gamma_{j'}^{\lambda}) , \tag{25}$$

$$\chi_2 = \frac{(m'\sqrt{S'} - m\sqrt{S})m\sqrt{S}}{\hbar V} \frac{\Gamma_0^\lambda}{\omega - \omega_\lambda} \sum_{i,j} (\Gamma_i^\lambda - \Gamma_j^\lambda)^* + \text{c.c.} \tag{26}$$

Using (8) the single sum in (26) becomes

$$\sum_{i,j} \rightarrow \left[\left[A_0^\lambda + \sum_{\delta_1} A_{\delta_1}^\lambda \right] (G_0^{(1)} - G_0^{(3)}) + \sum_{\delta_2} A_{\delta_2}^\lambda (G_0^{(2)} - G_0^{(4)}) \right]^* \tag{27}$$

and the double sum in (25) is

$$\sum_{\substack{ii' \\ jj'}} = \sum_{i,j} \left[\sum_{i,j} \right]^* .$$

From (27) one can see that the s modes are the only ones which respond to a uniform excitation, since only for them A_0, $\sum_{\delta_1} A_{\delta_1}$, and $\sum_{\delta_2} A_{\delta_2}$ are nonzero. For the s modes we can use (10), (15), and (24)–(27) to find the final result for the susceptibility. If we assume that there are cN impurities in the up sublattice and that their local modes are noninteracting, the total susceptibility becomes

$$\chi_T(\omega \cong \omega_s; 0) = \frac{cNm^2 S}{\hbar V} \left[\frac{2\Gamma_0^s \mu B(\omega)}{[(\omega_s + \omega_H)^2 - \omega_0^2]} + \frac{B(\omega)^2}{[(\omega_s + \omega_H)^2 - \omega_0^2]^2} \right] \frac{1}{\omega - \omega_s} , \tag{28}$$

where

$$\begin{aligned} \mu &= \frac{m'}{m} \left[\frac{S'}{S} \right]^{1/2} - 1 , \\ B(\omega) &= A_0^s \omega_E [(1 + 2a_1)(\omega^+ - 1) \\ &\qquad - 8a_2(\omega^- + 1)] . \end{aligned} \tag{29}$$

Thus we see that in the noninteracting impurity approximation the intensity of the s local mode response is proportional to the impurity concentration c and it increases rapidly as its frequency ω_s (which *decreases* with increasing field) approaches the *down-going* host magnon frequency $\omega_0 - \omega_H$. Note that the cN impurities in the down sublattice contribute to χ^{--} but not to χ^{++}. Therefore, χ^{--} is enhanced as the s-mode frequency that *increases* with increasing field approaches the *up-going* magnon frequency $\omega_0 + \omega_H$. This conclusion has been verified in the experiments with FeF_2:Mn. Conversely, in CoF_2:Mn there is an enhancement when either impurity mode approaches the lower magnon frequency, owing to the mixing of the two modes in this system.[26]

In the far-infrared absorption the radiation-spin coupling factor is $m = g\mu_B$, and the product $m'\sqrt{S'}$ for the impurity is close to that of the host, so that μ is small. In fact, for the FeF$_2$:Mn system $g'=2.0$, $S'=2.5$, $g=2.23$, $S=2$, and $\mu=-0.03$, so that the first term in the large parentheses in (28) is negligible. One can define the enhancement Λ of the impurity response as the ratio between (28) and the "isolated" impurity susceptibility

$$\chi_{\text{imp}} = cNg^2\mu_B^2 S/\hbar V(\omega-\omega_s)\ , \tag{30}$$

$$\Lambda = \frac{B(\omega)^2}{[\omega_0^2-(\omega_s+\omega_H)^2]^2}\ .$$

In the Raman scattering experiment the coupling of the radiation with the impurity and host spins can be quite different from each other. In fact, in the case of FeF$_2$:Mn the coupling with the impurity spins is negligible and $m'/m \cong 0$, so that $\mu \cong -1$. In this case there is no meaning in comparing the local mode and the isolated impurity intensities. Instead, a convenient quantity to be compared with the Raman scattering measurements is the intensity of the local mode *relative* to the host mode, normalized with respect to the impurity concentration c. Defining the ratio

$$\eta = \frac{[(\omega-\omega_s)\chi_T(\omega)]_{\omega_s}}{c[(\omega-\omega_0)\chi_0(\omega)]_{\omega_0}} \tag{31}$$

one obtains from (23) and (28)

$$\eta = \frac{\omega_0}{\omega_A}\left[\frac{2\Gamma_0^s\mu B(\omega)}{(\omega_s+\omega_H)^2-\omega_0^2} + \frac{B(\omega)^2}{[(\omega_s+\omega_H)^2-\omega_0^2]^2}\right]\ . \tag{32}$$

This parameter corresponds to the ratio of the areas under the Raman lines at ω_s and at ω_0, divided by c. It can be readily measured in inelastic light-scattering experiments[16] in which the line shapes do not suffer the distortions due to the effect of magnetic polaritons that are present in the case of the infrared absorption.[15]

Before proceeding to the numerical applications it is instructive to examine the results obtained so far in the limiting case where the s-mode frequency lies in the antiferromagnetic gap just below the $k=0$ magnon. In this case one can obtain simple analytical expressions for the wave functions and for the enhancement. When $\omega_0^2-\omega_s^2 \ll \omega_E^2$ the contribution of the center of the Brillouin zone dominates the expression for the Green's function (13). One can then use the parabolic approximation for the dispersion relation and replace the discrete sum in (13) by an integral,

$$U_{l0} \cong -\frac{4}{(2\pi)^3}\int d^3\tilde{k}\,\frac{e^{i\tilde{k}\cdot\vec{\mathrm{R}}}}{b^2+\tilde{k}^2}\ , \tag{33}$$

where $\vec{\mathrm{k}}\cdot\vec{\mathrm{r}}_l = \tilde{k}\cdot\vec{\mathrm{R}}_l$, $R_l=(l^2+m^2+n^2)^{1/2}$ is the normalized distance of the site l (coordinates la,ma,nc) to the origin, and b is a range parameter defined by

$$b = 2\left[\frac{\omega_0^2-\omega'^2}{\omega_E^2}\right]^{1/2}\ . \tag{34}$$

For $l \neq 0$ and b sufficiently small the upper limit of the integral can be extended to infinity and the Green's function takes the form of a Yukawa potential,

$$U_{l0} \cong -\frac{1}{\pi}\frac{e^{-bR_l}}{R_l}\ . \tag{35}$$

If for the sake of simplicity we assume that the nearest-neighbor impurity-host interaction J_1' is zero, $a_1=0$ and the local s-mode wave function (16) becomes approximately

$$\Gamma_{l\neq 0}^s \cong -A_0^s\frac{\omega^+}{\pi\omega_E}(1+8a_2\omega^-)\frac{e^{-bR_l}}{R_l}\ , \tag{36}$$

where $\xi = 1/b$ can be identified with the impurity mode radius, which varies with frequency as $(\omega_0^2-\omega'^2)^{-1/2}$. This dependence of the local mode disturbance on the distance to the impurity can also be obtained by a semiclassical treatment of the motion of the spins.[27] Actually it is found in other types of impurity problems in crystals[28] since the electron energy also varies as k^2 near the edge of the bands. As shown by Ivanov and Pogorelov[28] a substantial change in the spectrum of the excitations occurs when the impurity concentration is comparable to the value $c_0=(4\pi\xi)^{-3}$. For the FeF$_2$:Mn system at a field of 50 kOe, $\xi \cong 2.2$ and $c_0 \sim 8\times 10^{-3}$, and indeed impurity banding effects are observed at impurity concentrations as low as this value.[29]

One can also express the enhancement in terms of the local mode radius. When $(\omega_0-\omega_s) \ll \omega_E$ the term in S_{00} dominates the denominator in (17). With the same approximations used to derive (35) one can show that

$$S_{00} \cong \frac{\omega_E(\omega^{+2}-1)}{\pi(\omega_0^2-\omega'^2)^{1/2}}\ . \tag{37}$$

Using this frequency dependence of S_{00} in (17) and

(29), and neglecting the slow frequency dependences of a_1 and a_2 in (30), one finds that the enhancement varies with the third power of the radius $\Lambda \propto \xi^3$. Of course ξ^3 represents an effective volume occupied by each impurity, and one expects that a system with dimension d will have an enhancement $\Lambda \propto \xi^d$. This explains why the enhancement in one-dimensional magnets is smaller[30] than in three-dimensional ones. We should note at this point that the presence of a competing J'_1 interaction modifies the wave function considerably, and the simple form (36) does not provide a good description for the local mode. This will be shown in the numerical calculations of the next section.

IV. APPLICATION TO (FeMn)F_2

In this section we apply the previous results to find the local s-mode energies, wave functions, and dynamic response of the system $Fe_xMn_{1-x}F_2$ in the two impurity regime limits, $x \rightarrow 0$ and $x \rightarrow 1$. This system has now been extensively studied experimentally by various techniques but the lack of a detailed theory has thus far precluded the determination of their interaction parameters. Table I shows some of the relevant physical parameters of pure FeF_2 and MnF_2. The mixed $Fe_xMn_{1-x}F_2$ system was first studied throughout the whole concentration range by Enders *et al.*[31] by Fourier-transform far-infrared spectroscopy. Those authors observed the strong Mn defect mode in the gap of FeF_2 close to the magnon band which had been predicted theoretically by Tonegawa.[20] They found that as the Mn content increased the "defect" frequency gradually decreased to become the 8.7-cm^{-1} antiferromagnetic resonance mode in pure MnF_2. On the other hand, the host Fe mode in FeF_2:Mn increases in frequency as x decreases, to become the s_0 Fe impurity mode in MnF_2. This mode had previously been studied[5,32] by Weber and Johnson with infrared techniques, and by Oseroff and Pershan[4] with light scattering, who determined that its frequency was 94.8 cm^{-1}. The Mn impurity mode in FeF_2 was studied in greater detail by high-resolution far-infrared laser spectroscopy[15] and by light scattering[16] techniques, and its frequency at $H_0 = 0$ was determined to be 50.27 cm^{-1}. Several of these studies showed that contrary to pure FeF_2 and MnF_2, in the impure system the Mn-Fe nearest-neighbor interaction J'_1 is sizable at both ends of the concentration. Thus we have calculated the features of the Mn impurity mode in FeF_2 and of the Fe mode in MnF_2 allowing J'_1 to vary from -2 cm^{-1} (antiferromagnetic) to $+2$ cm^{-1} (ferromagnetic).

The Green's functions in Eq. (13) were calculated for frequencies outside the magnon band by transforming the sums in k into integrals and approximating them by coarse sums, which were evaluated in a PDP-10 computer. For frequencies close to the bottom of the band the dominant contribution to the sum comes from the center of the Brillouin zone, so the octant of the zone was divided into $30 \times 30 \times 30$ points and the center volume was further divided into $20 \times 20 \times 20$ points. For frequencies not close to the magnon band the finer sum in the center volume was found unnecessary. The Green's functions obtained with this procedure agree within four decimal digits with those tabulated by Walker *et al.*[33] Once the Green's functions are known, the frequencies, the wave functions, and the enhancement of the intensity of the s modes are calculated in a straightforward manner with Eqs. (14)–(18) and (28).

A. FeF_2:Mn

The calculations were made with the host parameters given in Table I and with the following impurity parameters: $\beta = 1.25$, $\delta' = 0.013$, $\omega_s = 50.27$ cm^{-1} (1.509 THz) at zero applied field and in the limit $c \rightarrow 0$. For each value of J'_1 in the range -2.0 to 2.0 cm^{-1} the value of the (antiferromagnetic) J'_2 necessary to fit the s-mode frequency was found with Eq. (14), and the result is shown in Fig. 1. Note that for $J'_1 = 0$ the value of J'_2 which fits the frequency is 1.74 cm^{-1}. These values correspond to an impurity mode Ising frequency of 56.58 cm^{-1}, which lies inside the magnon band 52.54–78.4 cm^{-1}. For $J'_1 \neq 0$ the discrepancy between the Ising value and the actual frequency is even larger (e.g., $J'_1 = -2.0$, $J'_2 = 2.5$ yield $\omega_I = 64.6$ cm^{-1}), which shows the inadequacy of the Ising model when the

TABLE I. Relevant physical quantities in MnF_2 and FeF_2 used in the calculations.

	S	g	ω_0 (cm^{-1})	ω_E (cm^{-1})	ω_A (cm^{-1})
FeF_2 (Refs. 15 and 40)	2	2.23	52.54	58.24	20.20
MnF_2 (Refs. 41 and 42)	2.5	2.0	8.69	48.80	0.77

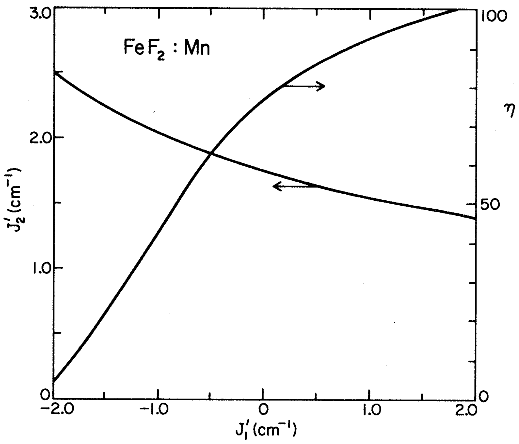

FIG. 1. Dependence of the impurity-host interaction parameter J_2' (antiferromagnetic) on the nearest-neighbor parameter J_1' (ferromagnetic if positive) to fit the frequency of the s_0 local mode in FeF$_2$:Mn to 50.27 cm^{-1}. Also shown is the variation of the relative Raman intensity, as defined in the text.

impurity mode is near the magnon band.

In order to determine the correct impurity-host interaction parameters one needs another independently measured quantity. The other two s-mode frequencies lie inside the magnon band and cannot be observed. At finite impurity concentrations there are several pair modes whose frequencies can be fitted to a two-impurity Green's-function theory.[34] A simpler measurement in this case is the relative intensity of the Raman impurity mode η, which is very sensitive to the value of J_1', as shown in Fig. 1. This was measured[16] for three concentration values, as shown in Fig. 2. Extrapolation to $c=0$ gives $\eta=75$, which corresponds to $J_1'=-0.2$ cm^{-1} and $J_2'=1.959$ cm^{-1}, both antiferromagnetic. The sign of J_1' agrees with Refs. 15 and 35. However, their conclusion that the magnitude is much larger than J_1 in the pure crystals FeF$_2$ and MnF$_2$ is not confirmed by our calculations. The cause of this discrepancy probably lies in the fact that when the local mode extends over many neighbors, the assignment of a magnetization to the impurity site masks the microscopic interaction parameters.

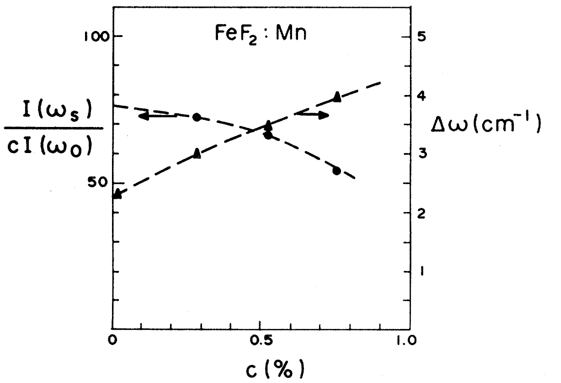

FIG. 2. Measured relative Raman intensity and frequency separation between the local and host modes in FeF$_2$:Mn for several impurity concentrations (Refs. 15 and 16).

The strong dependence of the Raman intensity on the value of J_1' (with fixed ω_s frequency) can be understood from the behavior of the wave functions of the impurity and its neighbors. Table II shows the wave functions $\Gamma_l^{(s)}$ of the s_0 mode at 50.27 cm^{-1}, obtained from Eq. (16), for several sites labeled by (l,m,n), which have position vectors $\vec{r}/a=l\hat{x}+m\hat{y}+c/an\hat{z}$ with respect to the impurity at $i=0$ (a and c are the lattice parameters). The parentheses under the indices indicate the number of equivalent sites and the last column gives the sum $S=\sum\Gamma_i^2-\sum\Gamma_j^2$ evaluated over a cluster with 97 spins, limited by the thirteenth-nearest neighbors. Notice that for a large antiferromagnetic interaction J_1', the nearest neighbor (001) and the impurity (000) wave functions have opposite phases. Since they are in the same spin sublattice, they produce competing interactions on the other neighbors, causing the phase of the wave function to oscillate from site to site. As the coupling between the radiation and the Mn^{2+} impurity spin is zero, the light scattering by the local mode is entirely due to the participation of the Fe^{2+} host spins around the impurity. Therefore, the response of the local mode decreases as J_1' increases antiferromagnetically (negative). It is interesting to note also that as the competition between J_1' and J_2' increases the local mode tends to spread out over a larger number of neighbors, as indicated by the smaller values of Γ_0 and of $\Gamma(\delta_2)$. In this case the local mode wave function is not described by the simple relation (36) obtained with $J_1'=0$ and its radius is larger than b^{-1}. So the antiferromagnetic interaction J_1' in FeF$_2$:Mn favors impurity-impurity interaction effects even at very small concentrations.

The effect of the magnetic field H_0 is to displace both the local mode and $k=0$ magnon frequencies downward in the case of an impurity in the up-spin sublattice. Since the g factor of iron is larger than that of manganese the frequency separation $\Delta\omega$ decreases as H_0 increases.[15] Figure 3 shows the behavior of $\Delta\omega$, the infrared absorption enhancement Λ, and the relative Raman intensity η as a function of H_0 in FeF$_2$:Mn. As expected, as $\Delta\omega$ de-

TABLE II. s_0 local mode wave functions at the impurity site and its near neighbors in FeF$_2$:Mn for several values of J'_1. In each case the value of J'_2 is the one that fits the frequency $\omega_s = 50.27$ cm^{-1}. The parentheses below give the number of equivalent sites. The last column shows the sum $S = \sum \Gamma_i^2 - \sum \Gamma_j^2$ computed in the cluster limited by the thirteenth-nearest neighbors ($r \sim 2a$).

Γ	(000)	(001)	$(\frac{1}{2}\frac{1}{2}\frac{1}{2})$	(100)	(101)	$(\frac{1}{2}\frac{1}{2}\frac{3}{2})$	(002)	(110)	
J'_1 cm^{-1})	(1)	(2)	(8)	(4)	(8)	(8)	(2)	(4)	S
−2.0	0.864	−0.324	0.075	0.096	0.046	−0.012	−0.013	0.063	0.986
−1.5	0.923	−0.176	0.085	0.112	0.063	0.002	0.006	0.075	0.971
−1.0	0.933	−0.050	0.090	0.120	0.074	0.013	0.021	0.081	0.958
−0.5	0.918	−0.049	0.092	0.123	0.081	0.022	0.032	0.084	0.949
−0.2	0.904	0.097	0.092	0.124	0.084	0.026	0.037	0.085	0.945
0	0.894	0.124	0.092	0.124	0.085	0.028	0.040	0.085	0.942
0.5	0.868	0.180	0.091	0.123	0.088	0.032	0.046	0.085	0.938
1.0	0.842	0.224	0.090	0.122	0.089	0.036	0.050	0.085	0.935
1.5	0.819	0.258	0.089	0.122	0.090	0.038	0.054	0.085	0.933
2.0	0.800	0.286	0.088	0.120	0.091	0.039	0.055	0.084	0.931

creases the local mode radius increases and its response to the external excitation is enhanced dramatically.

B. MnF$_2$:Fe^{2+}

Since the single-ion anisotropy of the Fe^{2+} impurity in this sytem is large, the value assigned to the parameter D' will have a strong effect on the values of J'_1 and J'_2 that fit the local mode frequency. Unfortunately, the values for D' used in the literature differ by as much as 20%. We have calculated J'_2 as a function of J'_1 which reproduce the s_0 local mode frequency at 94.8 cm^{-1} using $D' = 10.1$ cm^{-1} (Ref. 36) and 8.1 cm^{-1} (Ref. 37), $\beta = 0.8$, and the host parameters of Table I. The results are shown in Fig. 4. Also shown in the figure is the Ising model impurity frequency given by

$$\hbar\omega_I = g'\mu_B H'_A + 2Sz_2 J'_2 + 2Sz_1 J'_1 \ . \qquad (38)$$

The local mode wave functions at several neighbor sites for $D' = 10.1$ cm^{-1} and various pairs of the parameters J'_1 and J'_2 are given in Table III. The last column in this table is the sum $S = \sum_i |\Gamma_i|^2 - \sum_j |\Gamma_j|^2$ evaluated over the cluster limited by the fourth-nearest neighbors. Its proximity to 1 gives an indication of the localization of the mode. Table III shows that, as is well

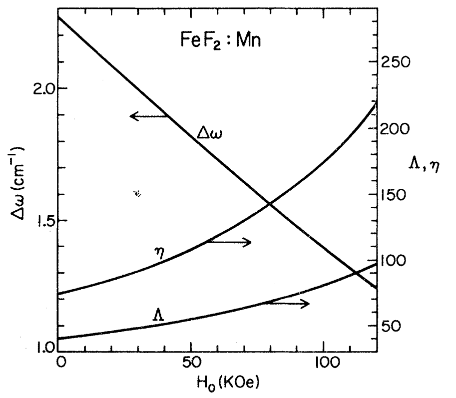

FIG. 3. Frequency separation to the host mode and intensity of the local s_0 mode in FeF$_2$:Mn as a function of the magnetic field applied along the c axis in the zero impurity concentration limit.

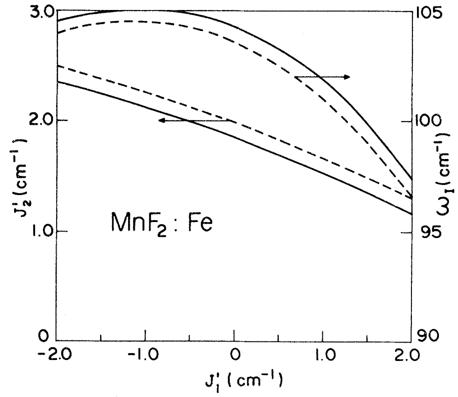

FIG. 4. J'_2 vs J'_1 that reproduce the s_0 mode in MnF$_2$:Fe at 94.8 cm^{-1}. The solid line corresponds to $D' = 10.1$ cm^{-1} and the dashed one to 8.1 cm^{-1}. Also shown is the impurity frequency obtained with the Ising approximation, Eq. (37).

TABLE III. s_0 local mode wave functions at the impurity site and its near neighbors in $\mathrm{MnF_2{:}Fe}$ for several values of J_1'. In each case the value of J_2' is the one that reproduces the frequency $\omega_s = 94.8\ \mathrm{cm}^{-1}$, with $D' = 10.1\ \mathrm{cm}^{-1}$. The parentheses below give the number of equivalent sites. The last column shows the sum $S = \sum \Gamma_i^2 - \sum \Gamma_j^2$ computed in the cluster limited by the fourth-nearest neighbors.

Γ	(000)	(001)	$(\frac{1}{2}\frac{1}{2}\frac{1}{2})$	(100)	(101)	$(\frac{1}{2}\frac{1}{2}\frac{3}{2})$	(002)	(110)	
J_1' (cm^{-1})	(1)	(2)	(8)	(4)	(8)	(8)	(2)	(4)	S
−2.0	0.997	0.130	0.069	−0.036	−0.018	0.003	−0.002	−0.017	0.998
−1.5	1.004	0.097	0.066	−0.034	−0.017	0.002	−0.001	−0.016	0.998
−1.0	1.008	0.059	0.062	−0.032	−0.016	0.001	0.000	−0.015	0.999
−0.5	1.010	0.018	0.057	−0.030	−0.014	−0.001	0.000	−0.014	0.999
0	1.008	−0.027	0.052	−0.027	−0.013	−0.002	0.001	−0.013	0.999
0.5	1.001	−0.076	0.046	−0.024	−0.011	−0.004	0.002	−0.011	0.999
1.0	0.989	−0.128	0.039	−0.020	−0.008	−0.006	0.003	−0.010	1.000
1.5	0.970	−0.183	0.032	−0.016	−0.006	−0.008	0.004	−0.008	1.000
2.0	0.955	−0.240	0.024	−0.012	−0.003	−0.010	0.005	−0.006	1.000

known, the impurity mode is very localized because its frequency lies well above the spin-wave band. However, the degree of localization is not high enough to allow one to neglect the wave function at the nearest and the next-nearest neighbors. As a result, though the Ising expression for the impurity mode frequency describes this system much better than $\mathrm{FeF_2{:}Mn}$, it departs from the Green's-function value as much as 10% in the range of variation of J_1' studied. The approximation becomes better as J_1' increases ferromagnetically because this favors the localization. Unfortunately the impurity mode response cannot be used to help in the determination of the impurity host parameters in this case. The relative (local)/(host) mode infrared absorption varies only from 1.3 to 1.5 in the range of Fig. 4. This is not surprising since the strong localization of the mode prevents the host neighbors from inducing an enhancement on the impurity spin response. The other measurements available in the literature on $\mathrm{MnF_2{:}Fe}$, such as the frequencies of several pair modes, determined by infrared[5,32,38] and Raman[4,39] spectroscopy, and the behavior of the magnetizations of the impurity and its neighbors, measured by NMR,[7] cannot be interpreted in the context of the theory presented here and so they are not to give an independent set of values of J_2' vs J_1'. But it is evident from Fig. 4 that our results are in disagreement with the values $J_1' = 0.7\ \mathrm{cm}^{-1}$, $J_2' = 1.42\ \mathrm{cm}^{-1}$ given in Ref. 4 and with $J_1' = 2.1\ \mathrm{cm}^{-1}$, $J_2' = 2.3\ \mathrm{cm}^{-1}$ given in Ref. 7. The former set gives energy values which are smaller than the actual energy both in the Green's-function calculation and in the Ising approximation, whereas the latter corresponds to too-large energy values.

V. SUMMARY

We have studied the features of magnetic impurity associated local modes in antiferromagnetic insulators in the limit of very small impurity concentrations and zero temperature using a Green's-function approach. With the theory developed we calculated the s_0 local mode frequencies, wave functions, and response to external excitation in $\mathrm{FeF_2{:}Mn^{2+}}$ and $\mathrm{MnF_2{:}Fe^{2+}}$. In the first system the local mode lies just below the spin-wave band of $\mathrm{FeF_2}$. As a consequence the mode spreads over many impurity neighbors and its dynamic response is enhanced dramatically with respect to the situation where the impurity is isolated. It is found that the degree of localization, and therefore the enhancement, depends strongly on the value of the nearest-neighbor impurity-host interaction parameter. Thus the data on the enhancement and the frequency of the impurity mode provide enough information to determine that $J_1' = 0.2\ \mathrm{cm}^{-1}$ and $J_2' = 1.79\ \mathrm{cm}^{-1}$, both antiferromagnetic. These values are in good agreement with the ones obtained by Belanger *et al.*[34] In $\mathrm{MnF_2{:}Fe^{2+}}$ on the other hand, the s_0 local mode lies well above the magnon band and is very localized. Thus the enhancement is small and the measurement of intensity of the impurity mode does not allow an unambiguous determination of the impurity-host interaction parameters.

ACKNOWLEDGMENTS

The author acknowledges many illuminating discussions with several colleagues with whom he worked on various aspects of this problem. In particular the interaction with Professor V. Jaccarino and Dr. R. W. Sanders of UCSB and Professor Belita Koiller, Professor Cid Araújo, and Professor Erivaldo Montarroyos was most beneficial. This work was supported in part by Financiadora de Estudos e Projetos (FINEP) and Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq).

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