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Dynamic response of local magnons: Single-impurity limit in one-dimensional magnets

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The dynamic response of local magnon modes associated with a single-impurity spin in one-dimensional ferro- and antiferromagnetic insulators is studied theoretically with the use of a Green's-function formulation solved exactly, by transfer-matrix techniques, for zero temperature. The calculations are applied to the typical one-dimensional (1D) ferromagnet $CsNiF_3$ and the antiferromagnet $(CH_3)_4NMnCl_3$ (TMMC) as functions of the impurity parameters in a way to allow the interpretation of possible future measurements of defect modes in these materials. The theory also explains qualitatively recent measurements in the three-dimensional defect antiferromagnets FeF_2:Mn^{2+}, CoF_2:Mn^{2+}, and FeF_2:Co^{2+}.

I. INTRODUCTION

The effects of substitutional impurities on the spin-wave modes in magnetic crystals have been under study for many years.[1] Many properties of these systems have been investigated theoretically and experimentally and are now well understood. The systems mostly studied are antiferromagnetic insulators in which the impurity modes lie far above the spin-wave band. In this case the wave functions are well localized around the defect and a number of approximate theories describe satisfactorily the properties of the modes. On the other hand, when the defect modes have frequencies very close to the magnon band, the localization of the wave functions is weak and a number of interesting effects occur. Recent far-infrared (FIR) laser experiments have shown for example that the response of the s_0 local modes of Mn impurities in CoF_2,[2] and FeF_2,[3] whose frequencies lie just below the spin-wave band, is largely enhanced by the interaction with the excitations of the host crystal. Similar experiments[4] with Co impurity modes in FeF_2 show that the modes close to the top of the band decrease in intensity as they move to lower energies with increasing magnetic fields. The behavior of the intensity in the first group of experiments has been explained satisfactorily by a semiclassical equations of motion approach,[3] but a more refined theory is needed to explain quantitatively the experimental data.

In this paper we develop an exact Green's-function calculation of the local mode response in one-dimensional (1D) magnets in the single impurity limit. Though the theory does not apply to the three-dimensional crystals studied experimentally so far, many results of the model can be used to explain the experiments qualitatively.

The usual theoretical approach to the one-impurity problem involves t-matrix techniques which require explicit calculation of the pure host crystal Green's function.[5–8] Here we treat the problem of a magnetic chain with one impurity within a transfer-function formulation[9,10] which does not involve knowledge of the pure lattice Green's function, and turns out to be much simpler than the usual treatments.[11–13] In Sec. II we consider the ferromagnetic case: the formalism for the calculation of the Green's function is presented and the local modes frequencies and wave functions are calculated. The conditions for existence of such modes are also obtained. In Sec. III we calculate the dynamic response of the system for optical excitation and neutron scattering at zero temperature. The antiferromagnetic chain is considered in Sec. IV, and our conclusions are presented in Sec. V.

II. FERROMAGNETIC HOST

We consider a chain of spins $\vec{S}_i$ described by the Hamiltonian:

$$\mathcal{H} = -\sum_{i,j} J_{ij}\vec{S}_i\cdot\vec{S}_j - \sum_i \gamma_i H_A^i S_i^z \ , \tag{1}$$

where $\gamma_i = \mu_B g_i$; the host crystal magnetic ions have spin S, nearest-neighbor ferromagnetic isotropic exchange constant J, and anisotropy field H_A. The single impurity at site $i=0$ has parameters S',J', and H_A'.

Let G_{ij} be the Fourier transform of the double-time Zubarev[14] Green's function for the *defect lattice:*

$$G_{ij} = G(i,j|\omega) = (4S_iS_j)^{-1/2} \ll S_i^+, S_j^- \gg_\omega \ . \tag{2}$$

The equation of motion for G_{ij} at zero temperature is

readily derived[9]:

$$(\omega-\omega_i^I)G_{ij}=\delta_{ij}-\sum_{j'}\omega_{ij'}^T G_{j'j} \ , \tag{3}$$

where

$$\omega_i^I=\sum_{j'\neq i}2\hbar^{-1}J_{ij'}S_{j'}+\gamma_i H_A^i \tag{4}$$

is the Ising frequency, and

$$\omega_{ij'}^T=2\hbar^{-1}J_{ij'}(S_iS_{j'})^{1/2} \tag{5}$$

is the transverse coupling frequency.

We are interested in the trace of G, which is associated with the density of magnon modes. The equations related to the diagonal element of the Green's function relative to the defect site are:

$$\begin{aligned}
&(\omega-\omega_0^I)G_{00}=1-2\omega_{01}^T G_{10} \ , \\
&(\omega-\omega_1^I)G_{10}=-\omega_{10}^T G_{00}-\omega_T G_{20} \ , \\
&(\omega-\omega_I)G_{20}=-\omega_T G_{10}-\omega_T G_{30} \ , \qquad (6)\\
&\qquad\vdots \\
&(\omega-\omega_I)G_{n0}=-\omega_T G_{n-1,0}-\omega_T G_{n+1,0} \ , \quad n\geq 2 \ , \qquad (7)
\end{aligned}$$

where

$$\omega_I=\omega_{n>1}^I=\frac{4JS}{\hbar}+\gamma H_A \ , \tag{8}$$

$$\omega_T=\omega_{n,n\pm1\neq 0}^T=\frac{2JS}{\hbar} \ . \tag{9}$$

This infinite set of coupled equations can be solved by the introduction of the transfer function

$$T(\omega)=\frac{G_{n,0}}{G_{n-1,0}}, \quad n\geq 2 \ , \tag{10}$$

which is obtained by using Eq. (10) in Eq. (7):

$$T(\omega)=\pm(\epsilon^2-1)^{1/2}-\epsilon \ , \tag{11}$$

with

$$\epsilon=(\omega-\omega_I)/2\omega_T \ . \tag{12}$$

The sign of the square root in Eq. (11) must be chosen to satisfy $\lim_{\omega\to\pm\infty}T(\omega)=0$, so that for $|\epsilon|>1$, the sign is the same as the sign of ϵ. For $|\epsilon|<1$ the sign must be chosen so that the density of states is positive in that range. Substituting Eq. (11) into Eq. (6), we get:

$$G_{00}=\frac{\omega-\omega_1^I+\omega_T T}{(\omega-\omega_0^I)(\omega-\omega_1^I+\omega_T T)-2(\omega_{01}^T)^2}\equiv\frac{B(\omega)}{C(\omega)} \ . \tag{13}$$

Analogously we obtain any element of the Green's function; in particular the diagonal element relative to the impurity nearest neighbor is:

$$G_{11}=\frac{(\omega-\omega_0^I)B(\omega)-(\omega_{01}^T)^2}{B(\omega)C(\omega)} \ . \tag{14}$$

The normalized local density of magnon states at site i is given by:

$$\rho_i(\omega)=-\pi^{-1}\,\mathrm{Im}G_{ii}(\omega)+\sum_\lambda R_{ii}^\lambda\delta(\omega-\omega_\lambda) \ , \tag{15}$$

where the sum is over the poles of G_{ii} and R_{ii}^λ is the residue of $G_{ii}(\omega)$ at a pole ω_λ. Note that the transfer function (11) is complex for ω in the interval $(\omega_I-2\omega_T, \omega_I+2\omega_T)$, or $-1<\epsilon<1$, which is the frequency range of propagating spin waves, thus in this range the density of states is a continuum. Localized impurity modes correspond to the poles of G_{ii}. Symmetric or s-like modes, i.e., modes with nonzero amplitude at the impurity site, appear as poles of G_{00}, while antisymmetric or p-like modes must be investigated at a different i site, G_{11} for example.

From Eqs. (13) and (14), we conclude that the zeros of $C(\omega)$ correspond to s-like mode frequencies, while the zeros of $B(\omega)$ correspond to p-like modes: these local mode frequencies are identical to the results of White and Hogan[11] and Tonegawa.[12] Conditions for the appearance of such modes above and below the continuum may be easily investigated from the analytic expressions for $B(\omega)$ and $C(\omega)$. These conditions are summarized in Table I, where we introduce for convenience the parameters $\alpha=J'/J$, $\beta=S'/S$, and $\delta=(g'\mu_B H_A'-g\mu_B H_A)/2JS$; explicit use is made of the fact that $\alpha>0$ and $\beta>0$ in order to simplify some of the conditions.

In Fig. 1 the frequencies of the s and p modes as a function of α are presented for several values of δ, keeping $\beta=2$, which applies to Fe^{++} impurities in the 1D anisotropic ferromagnet $CsNiF_3$. Note that for negative values of the relative anisotropy parameter δ an s_0 mode appears below the magnon band. Modes of this type have been observed in the 3D antiferromagnets CoF_2:Mn,[2,15] and FeF_2:Mn.[3,15] The s_1 mode appears above the magnon band. s-like modes have been observed in many 3D antiferromagnets,[1] but only recently experiments were reported[4] with modes of this type very close to the top of the band.

TABLE I. Conditions for the existence of localized magnon modes in a ferromagnetic chain with one impurity: $\alpha=J'/J$, $\beta=S'/S$, and $\delta=(\gamma'H_A'-\gamma H_A)/2JS$.

	Above the continuum	Below the continuum
s-like mode	$(4-\delta)(\alpha\beta-2)>4\alpha$	$\delta<0$
p-like mode	$\alpha\beta>2$	...

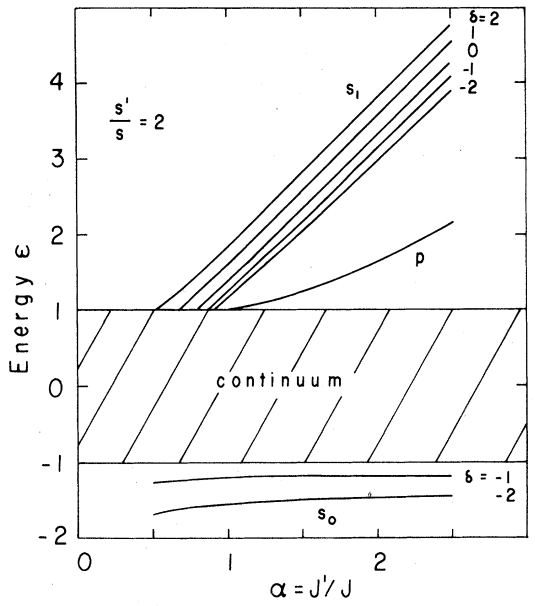

FIG. 1. Frequencies $\epsilon=(\omega-\omega_I)/2\omega_T$ of the s and p modes for a ferromagnetic linear chain as functions of the exchange constants for various values of the anisotropy parameter $\delta=(\gamma' H_A'-\gamma H_A)/2JS$.

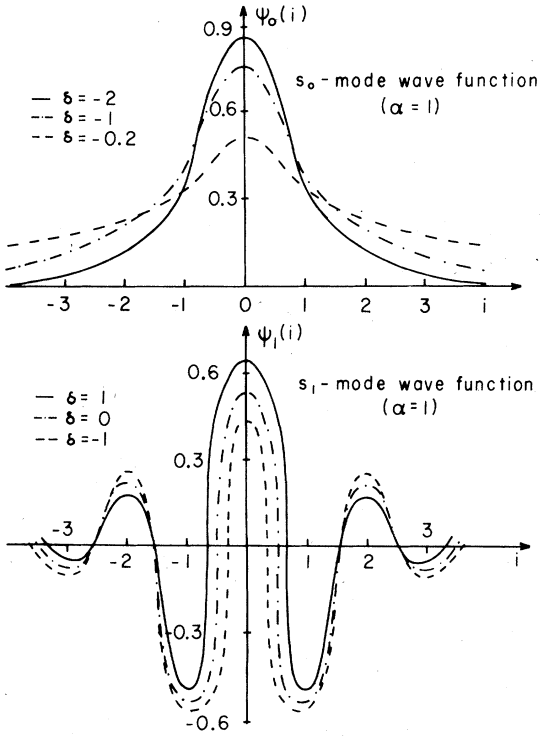

FIG. 2. Spin-deviation amplitudes around the impurity site ($i=0$) for the s_0 and s_1 local magnon modes for $\alpha=1$ and $\beta=2$.

The wave function of the local modes may be easily obtained from the residues of the poles corresponding to these modes for different elements of the Green's function[16]:

$$|\psi_\lambda(i)|^2=R_{ii}^\lambda \ , \tag{16}$$

$$\frac{\psi_\lambda(i)}{\psi_\lambda(j)}=\frac{R_{ij}^\lambda}{R_{jj}^\lambda} \ . \tag{17}$$

The spin deviation amplitudes, $\psi_\lambda(i)$, at the vicinity of the impurity are plotted in Fig. 2 for $\alpha=1$, $\beta=2$, and different values of δ. Of course, only the values corresponding to integer i are physically meaningful. Notice that the s_0 mode corresponds to an excitation of the impurity spin in phase with its neighbors, while in the s_1 mode the impurity spin is in opposite phase with respect to its neighbors. This symmetry aspect is important in the dynamic excitation of these modes treated in Sec. III.

From Fig. 2 it is clear that the modes nearer to the band edges are less localized, and in this situation the Ising approximation gives very poor description of the local mode frequency. Modes with frequencies far from the continuum become Ising-like and are well localized at the impurity.

III. DYNAMIC RESPONSE

The response of the spin system to $k\simeq 0$ electromagnetic radiation such as used in Raman scattering or infrared-absorption experiments can be described by the susceptibility, written in terms of the Green's function as[1]:

$$\chi(\omega)=\sum_{ij}\frac{2(S_iS_j)^{1/2}}{V}\gamma_i\gamma_j G_{ij}(\omega) \ . \tag{18}$$

Due to the property of the Green's-function matrix elements to be related to each other by powers of the transfer function (except G_{00}), the summation in Eq. (18) can be grouped into geometric series, and therefore can be performed exactly. For frequencies near a pole of G, i.e., near a local mode frequency ω_λ, the susceptibility is:

$$\chi_\lambda(\omega)\simeq\frac{A}{\omega-\omega_\lambda}\left[\frac{1+T(\omega)[1-2R_{00}^\lambda]+4R_{10}^\lambda}{1-T(\omega)}+\frac{2R_{1,1}^\lambda}{[1-T(\omega)]^2}\right] , \tag{19}$$

where $A=2S\gamma^2/V$; use was made of the sum rule $\sum_{i=-\infty}^{\infty}R_{ii}^\lambda=1$ and for simplicity it was assumed $S'\gamma'^2=S\gamma^2$.

Notice that $\chi_{\text{imp}}=A/(\omega-\omega_\lambda)$ is the susceptibility of one isolated impurity spin; therefore the lattice-impurity spin interaction may change drastically the response of the local mode to the external field.

TABLE II. Reduced energy $\epsilon=(\omega-\omega_I)/2\omega_T$ of the s_0 and s_1 local modes, relative $k=0$ dynamic response $\eta=\chi_\lambda(\omega)/\chi_{\rm imp}$ and neutron scattering form factor $\xi=S_\lambda(\pi/a,\omega)/S_{\rm imp}$ as a function of δ for $\alpha=1$ and $\beta=2$.

	δ	ϵ	η	ξ
s_0	−2	−1.54	3.65	1.82
	−1	−1.20	5.84	1.60
	−0.2	−1.01	21.86	0.74
	−0.1	−1.004	41.54	0.45
s_1	−10	1.04	0.17	13.12
	−8	1.05	0.18	10.96
	−6	1.08	0.19	8.74
	0	1.41	0.05	1.71
	6	3.35	0.27	0.14

Since the value of the transfer function (11) at the uniform mode ($k=0$ magnon frequency) is $T(\omega_{k=0})=1$, the response of the local mode is largely enhanced by its proximity to the bottom of the magnon band. Local mode enhancements of the order of 100 have been observed in CoF_2:Mn (Ref. 2) and in FeF_2:Mn (Ref. 3), where the Mn mode frequency is just below the magnon band. This effect makes the impurity mode as intense as the uniform resonance mode for Mn concentrations of the order of 0.1%. The ratio $\eta=\chi_\lambda/\chi_{\rm imp}$ for the s_0 and s_1 modes calculated from Eq. (19) are presented in Table II. Notice that while the s_0 mode is enhanced by the impurity-lattice interaction, the s_1 mode is greatly weakened as it approaches the magnon band, as observed in FeF_2:Co.[4] This effect can be understood as resulting from the action of the transverse exchange field of the neighbors at the impurity spin: the s_0 mode is in phase with the $k=0$ extended mode, while the s_1 mode is not, so that the excitation of the neighbors at sites ±1 by the uniform external field partly cancels the excitation of the impurity mode with s_1 symmetry.

While the s_0 mode response is enhanced as its frequency approaches the $k=0$ magnon frequency, it is not as large as it would be for a 3D spin system with the same local mode frequency relative to the magnon band and same exchange constants. This is in part due to the smaller number of neighbors and consequently smaller exchange field of the 1D system.

The inelastic neutron scattering cross section by one spin-wave excitation in the chain is related to the dynamic structure factor[1,9]:

$$S(k,\omega)=\frac{1}{N}\sum_{ij}(S_iS_j)^{1/2}b_ib_j\,e^{ika(i-j)}G_{ij}(\omega)\quad,\quad(20)$$

where a is the lattice parameter and b_i is an interaction constant. Assuming for simplicity $S'b'^2=Sb^2$, expression (20) may be evaluated in the same way as Eq. (18); for frequencies near a local mode frequency ω_λ we get:

$$S_\lambda(k,\omega)=\frac{B}{\omega-\omega_\lambda}\,\mathrm{Re}\left[\frac{1+T(\omega)(1-2R^\lambda_{00})e^{ika}+4R^\lambda_{10}}{1-T(\omega)e^{ika}}+2R^\lambda_{-1,1}\left(\frac{e^{ika}}{1-T(\omega)e^{ika}}\right)^2\right],\quad(21)$$

which for $k=0$ is analogous to Eq. (19). The form factor for one isolated impurity spin is $S_{\rm imp}=B/(\omega-\omega_\lambda)$. Values of the ratio $\xi_\lambda(k)=S_\lambda(k,\omega)/S_{\rm imp}$ with $k=\pi/a$ for the s_0 and s_1 modes are given in Table II. Notice that the ratio $\xi_{s1}(\pi/a)$ is enhanced for ω_{s1} approaching the top of the magnon band.

While in light scattering experiments the response of local modes with symmetry other than s_0 is always weakened, the present results encourage the observation of such modes in neutron scattering experiments, even at very low concentrations.

IV. ANTIFERROMAGNETIC HOST

The calculations of the previous sections can be readily extended to an antiferromagnetic linear chain with one impurity. We assume a Hamiltonian describing the system as:

$$H=\sum_{ij}J_{ij}\vec{S}_i\cdot\vec{S}_j-\sum_i\gamma_iH^i_A\alpha_iS^z_i\quad,\quad(22)$$

where $\alpha_i=1$ if $\vec{S}_i$ belongs to the "up" sublattice and $\alpha_i=-1$ if $\vec{S}_i$ belongs to the "down" sublattice. The

other parameters are as defined in Sec. II (notice that the exchange coupling constant is positive here). The one-magnon Green's functions are defined as[1,10]:

$$G_{ij}^{\alpha\beta}(\omega) = G^{\alpha\beta}(i,j|\omega) = (-1)^{(1-\alpha)/2}(4S_iS_j)^{-1/2}\langle\langle S_i^+(\alpha), S_j^-(\beta)\rangle\rangle_\omega \ , \tag{23}$$

where the notation indicates that $\vec{S}_i$ belongs to sublattice α and $\vec{S}_j$ belongs to sublattice β.

Assuming the Néel ground state, the equations of motion for zero temperature are:

$$(\omega - \alpha\omega_i^l)G_{ij}^{\alpha\beta} = \delta_{ij} + \sum_{j'}(-1)^{(1+\alpha)/2}\omega_{ij'}^T G_{j'j}^{\bar{\alpha}\beta} \ , \tag{24}$$

where $\bar{\alpha} = -\alpha$ and ω_i^l, ω_{ij}^T are defined in Eqs. (4) and (5). Equation (24) may be solved by defining two transfer functions:

$$T_1(\omega) = \frac{G_{ij}^{-+}(\omega)}{G_{(i-1),j}^{++}(\omega)} = \frac{(\omega^2-\omega_l^2) \pm [(\omega^2-\omega_l^2)(\omega^2-\omega_l^2+4\omega_T^2)]^{1/2}}{2\omega_T(\omega+\omega_l)} \ , \tag{25}$$

$$T_2(\omega) = \frac{G_{ij}^{++}(\omega)}{G_{(i-1),j}^{-+}(\omega)} = -\frac{\omega+\omega_l}{\omega-\omega_l}T_1(\omega) \ . \tag{26}$$

Notice that the transfer functions, and therefore G, are complex for ω in the interval

$$[\omega_c = (\omega_l^2 - 4\omega_T^2)^{1/2}, \omega_l] \ ,$$

which is the frequency band of antiferromagnetic magnons. We solve Eq. (24) for the diagonal elements of the Green's function at the impurity site and at its nearest neighbor:

$$G_{00}^{++}(\omega) = G_{00}^{--}(-\omega) = \frac{\omega+\omega_1^l+\omega_T T_2(\omega)}{(\omega-\omega_0^l)[\omega+\omega_1^l+\omega_T T_2(\omega)]+2(\omega_{10}^T)^2} \ , \tag{27}$$

$$G_{11}^{++}(\omega) = G_{11}^{--}(-\omega) = \frac{(\omega+\omega_0^l)(\omega-\omega_1^l-\omega_T)T_1(\omega)+(\omega_{10}^T)^2}{(\omega-\omega_1^l-\omega_T)T_1(\omega)[(\omega+\omega_0^l)(\omega-\omega_1^l-\omega_T T_1)+2(\omega_{10}^T)^2]} \ . \tag{28}$$

The local density of states is obtained from Eq. (15) with $G_{ii}(\omega)$ taken as $[G_{ii}^{++}(\omega)+G_{ii}^{--}(\omega)]$. The conditions for existence and symmetry of the localized magnon modes are easily obtained from Eqs. (27) and (28); the results are summarized in Table III.

Figure 3 shows the frequencies of the local modes as a function of $\alpha = J'/J$ for $\beta = S'/S$ equal to 1/2.5 and to 2/2.5 for various values of the impurity anisotropy parameter $\delta' = \gamma' H_A'/2JS$. The results apply to the typical linear chain antiferromagnet $(CH_3)_4NMnCl_3$ (TMMC), for which the anisotropy energy is small (we have taken $H_A = 0$), and the two values of the spin considered apply to Ni^{++} or Fe^{++} ions substituting a Mn^{++} ion.

The dynamic response of the antiferromagnetic system is calculated in the same way as the ferromagnetic one, and the same qualitative behavior is obtained. Since the anisotropy of TMMC is very small, the magnon gap is small and no localized s_0 mode is expected to exist, therefore no enhancement effect is expected to be observed for this mode. However an enhancement in the neutron scattering by s_1 modes just above the band is expected to occur.

TABLE III. Conditions for the existence of localized magnon modes in an antiferromagnetic chain with one impurity: $\alpha = J'/J$, $\beta = S'/S$, $\delta' = \gamma' H_A'/2JS$, $\gamma = \omega_c/\omega_T$.

	Above the continuum	Below the continuum
s-like mode	$\alpha - 1 > \frac{1}{2}(\delta - \delta')$	$\alpha\beta(\gamma-\delta') - \alpha(\delta+\gamma) > \frac{1}{2}(\delta'-\gamma)(\delta+\gamma)$
	or	or
	$(\alpha\beta-1)(2+\delta+\delta') > 2\alpha$	$\alpha\beta(\delta'+\gamma) - \alpha(\gamma-\delta) > \frac{1}{2}(\delta'+\gamma)(\gamma-\delta)$
p-like mode	$\alpha\beta > 1$	$\delta(1-\alpha\beta) > \alpha^2\beta^2$

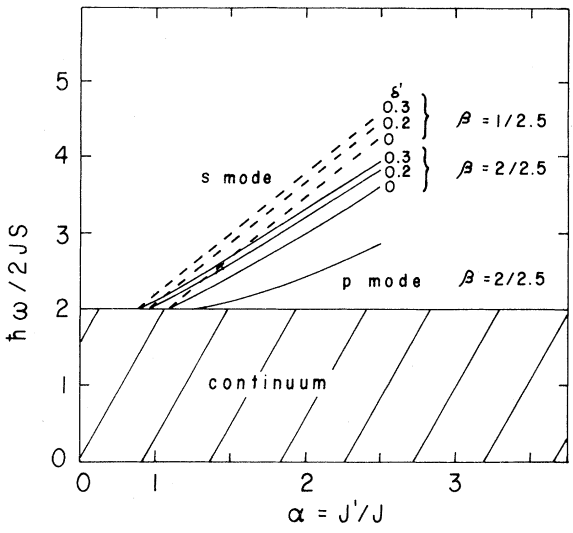

FIG. 3. Frequencies of the s_1 and p local modes in an antiferromagnetic linear chain as functions of the exchange constants and impurity anisotropy $\delta' = \gamma' H_A'/2JS$, for two values of the impurity spin. The anisotropy of the host spins is taken to be zero.

V. CONCLUSIONS

In this work we have presented a transfer-function treatment for the one-impurity problem in one-dimensional magnetic systems, which is very suitable for the calculation of the local mode energies and dynamic response. For impurity concentrations such that impurity-impurity interactions can not be neglected, the formalism may be extended in the alloy transfer-matrix approximation.[17,18] Such a calculation is in progress, and information about line position, shape, and widths may be obtained.

The observation of local magnon modes in one-dimensional ferro- and antiferromagnetic materials should not be much more difficult than in the 3D materials extensively explored. All the techniques used to study magnetic defects, Raman and neutron scattering, far infrared laser spectroscopy, and NMR are appropriate for these observations. The feasability of the preparation of 1D systems with magnetic impurities has been reported for TMMC crystals doped with Cu, Co, and Ni.[19] These systems would be strong candidates for the suggested observation. The fact that the ordering temperature of these systems is low presents some complications, but these are not major ones. In fact, these measurements should be possible even at temperatures above the ordering temperature, as long as the spin-spin correlation length is larger than the spread of the local mode. Since this spread accounts for only a few lattice spacings, depending on the proximity to the magnon band, local magnons may exist in TMMC at temperatures as high as 10 K.

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