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Localized and virtual magnetoelastic states in a linear chain

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1. INTRODUCTION

CONSIDERABLE attention has been given in the last few years to the effect of magnetic and elastic impurities, respectively, on the dynamics of spins in ferromagnetic systems, and on lattice dynamics. This is due to their influence on such diverse phenomena as thermal conduction, neutron scattering, Mösbauer effect, and others, in the materials in which they are present.

On the other hand, the magnetoelastic interaction and its consequences on magnon and phonon dynamics have been treated by Kittel[1], Akhiezer *et al.*[2], and on its bearing on thermal conductivity, by Erdös[3].

Recently, Stojanović and Tósić have considered the effect of the m.e. interaction on surface and volume states[4].

Our purpose in this paper is to study the effect of the m.e. coupling on the dynamics of a linear chain in which, the harmonic approximation has been taken for the elastic properties, and whose magnetic behavior is described with the Heisenberg model in the one magnon approximation.

Some intrinsic qualitative conclusions regarding the effect of the impurities and the m.e. interaction are directly extensible to the case of three dimensional lattices, since the same calculation techniques may be employed in both cases. As an example, the conclusion that the m.e. interaction turns, under certain conditions, a localized state into a virtual one, which follow in the discussion on scattering states, is valid both for the one dimensional and the three dimensional cases.

To our knowledge, in the extensive literature on the properties of localized and virtual states due to impurities in an otherwise perfect crystal, this part of the Hamiltonian has never been taken into account. The effect of magnetic impurities on the behavior of spins systems has been treated in[5–12], while the effect of impurities on the dynamics of the lattice is described in[13–21].

2. FORMULATION OF THE PROBLEM IN THE LINEAR CHAIN MODEL

The Hamiltonian we choose to describe the system is:

$$H = H_p + H_m + H_I \tag{1}$$

with

$$H_p = H_p^\circ + H_p' \tag{1a}$$

$$H_m = H_m^\circ + H_m' \tag{1b}$$

$$H_I = H_I^\circ + H_I'. \tag{1c}$$

H_p is the Hamiltonian describing phonons. In (1.1) it is split into an unperturbed part describing free elastic waves (H_p°), and a perturbing term due to the impurity (H_p').

In terms of the displacements from equilibrium and their canonically conjugate momenta they are:

$$H_p^\circ = \frac{1}{2}\sum_i \frac{p_i^2}{m} + r(x_i - x_{i-1})^2 \tag{2a}$$

$$H_p' = \frac{\mu m}{2}\dot{x}_0^2 + \frac{m\nu}{2}[(x_1 - x_0)^2 + (x_0 - x_{-1})^2]$$

where m and r are the constants of the perfect chain and

$$\mu=\frac{m'-m}{m}\qquad \nu=\frac{r'-r}{m}.$$

Here m', r' are the constants characterizing the impurity.

The Heisenberg Hamiltonian, using the Holstein–Primakoff transformation [22]

$$a_i=\frac{S_{xi}+iS_{yi}}{\sqrt{2S}}\qquad a_i^*=\frac{S_{xi}-iS_{yi}}{\sqrt{2S}}$$

can be written (to terms of second order in aa^*) in the form:

$$H_m^\circ=(2JSz+g\beta H)\sum_j a_j^*a_j-JS\sum_j (a_j^*a_{j+1}+a_{j+1}^*a_j+a_j^*a_{j-1}+a_{j-1}^*a_j)\quad (2b)$$

and

$$H_m'=2Sz(J'-J)a_0^*a_0+2(J'S'-JS)\times(a_1^*a_1+a_{-1}^*a_{-1}).\quad (2c)$$

In the foregoing, quantities related to the impurity are primed (we have used the same notation as Ref. [5]). Finally we have chosen for the magnetoelastic Hamiltonian:

$$H_I^\circ=-\frac{iV}{\sqrt{2}}\sum_j (a_j-a_j^*)(2x_j-x_{j+1}-x_{j-1})\quad (2e)$$

and

$$H_I'=-\frac{i(V'-V)}{\sqrt{2}}(a_0-a_0^*)(2x_0-x_1-x_{-1})\quad (2f)$$

where V is a parameter giving the strength of the magnetoelastic coupling.

We use Poisson brackets (like $\{S_x,S_y\}=iS_z/\hbar$) to calculate the time derivatives of a_j, a_j^* and x_j, and obtain:

$$i\hbar\dot{a}_l=-(\omega_0+2\omega_1)a_l+\omega_1(a_{l+1}+a_{l-1})-\frac{iV}{\sqrt{2}}(2x_l-x_{l+1}-x_{l-1})-2\epsilon\omega_1a_0\delta_{l,0}-\rho\omega_1(a_1\delta_{l,1}+a_{-1}\delta_{l,-1})+\gamma\omega_1(\delta_{l,1}+\delta_{l,-1})a_0+\gamma\omega_1(a_1+a_{-1})\delta_{l,0}-\frac{i}{\sqrt{2}}(V'-V)\delta_{l,0}(2x_0-x_1-x_{-1})\quad (3a)$$

$$-i\hbar\dot{a}_l^*=-(\omega_0+2\omega_1)a_l^*+\omega_1(a_{l-1}^*+a_{l-1}^*)+\frac{iV}{\sqrt{2}}(2x_l-x_{l+1}-x_{l-1})-2\epsilon\omega_1a_0^*\delta_{l,0}-\rho\omega_1(a_1^*\delta_{l,1}+a_{-1}^*\delta_{l,-1})+\gamma\omega_1(\delta_{l,1}+\delta_{l,-1})a_0^*+\gamma\omega_1(a_1^*+a_{-1}^*)\delta_{l,0}+\frac{i}{\sqrt{2}}(V'-V)\delta_{l,0}(2x_0-x_1-x_{-1})\quad (3b)$$

$$\ddot{x}_l=\frac{r}{m}(x_{l+1}+x_{l-1}-2x_l)+\frac{i}{\sqrt{2}}\frac{V}{m}2(a_l-a_l^*)-(a_{l+1}-a_{l+1}^*)-(a_{l-1}-a_{l-1}^*)-\mu\ddot{x}_0\delta_{l,0}-\nu(x_1-x_0)(\delta_{l,1}-\delta_{l,0})-\nu(x_{-1}-x_0)(\delta_{l,-1}-\delta_{l,0})+\frac{i(V'-V)}{\sqrt{2}}(a_0-a_0^*)(2\delta_{l,0}-\delta_{l,1}-\delta_{l,-1})\quad (3c)$$

where

$$\omega_0=g\beta H_z,\qquad \omega_1=2JS.$$

The effect of H_I° is to mix the elastic and magnetic running wave modes. Interference effects make this mixing negligible for all frequencies not close to those satisfying $\omega_m(q) = \omega_p(q)$ (see (1, 2)).

In the following, we shall neglect H_I°, so that our solutions will be valid for frequencies not near the resonance condition. The time Fourier transforms of a, a^* and x satisfy the following equations (in matrix form):

$$M^+a = V^M a + V^{MP} x \quad (4a)$$

$$M^- a^* = V^M a^* + V^{MP} x \quad (4b)$$

$$Lx = V^P x + V^{PM} a + V^{PM*} a^* \quad (4c)$$

a, a^*, x now indicate vectors whose j-th component is a_j, a_j^*, x_j; the matrices M^+, M^-, ν^M, etc. follow easily from equations (3).

Using the free magnon and phonon Green's functions G_0 and F_0 ((23, 5, 6, 7, 8, 9, 13, 15, 17) and others), these equations can be written in the following way:

$$a = a_0 + G_0^+ V^M a + G_0^+ V^{MP} x \quad (5a)$$

$$a^* = a_0^* + G_0^- V^M a^* + G_0^- V^{*MP} x \quad (5b)$$

$$x = x_0 + F_0 V^P x + F_0(V^{PM} a + V^{*PM} a^*) \quad (5c)$$

where a_0, a_0^* and x_0 are the solutions of the homogeneous system. (They are equal to zero if ω is outside the bands).

From equation (5) the solutions for different values of ω may be derived.

Several cases arise depending upon whether ω lies outside or inside the bands:

(a) *ω outside the magnon and phonon bands. (Localized states)*

In this case, it is more convenient to consider the subvectors A, A^* and X, formed by the -1, 0, 1 components of a, a^* and x. Since the V matrices have non zero elements only in this subspace, we can obtain from equation (5) equations involving only A, A^* and X. These being a subset of those equations, they have the same form, except that vectors and matrices must now be considered in the subspace $(-1, 0, 1)$. In order that this system have non trivial solutions it is necessary that either:

$$D_M(\omega) = \det\{1 - G_0^+(V^M + V^{MP} F V^{PM} + V^{MP} F V^{PM} G_{(1)}^- V^{MP} F V^{PM}\} = 0 \quad (6a)$$

or

$$D_P(\omega) = \det\{1 - F_0(V^P + V^{PM} G_+ V^{MP} + V^{PM*} G_- V^{MP*})\} = 0 \quad (6b)$$

where

$$G_{(1)}^{\pm} = (1 - G_0^{\mp} V_{\text{eff}}) G_0^{\mp}$$

$$V_{\text{eff}}^+ = V^M + V^{MP} F V^{PM}$$

$$F = (1 - F_0 V^P)^{-1} F_0$$

$$G^{\pm} = (1 - G_0^{\mp} V^M)^{-1} G_0^{\pm}.$$

Equations (6a) are exact and equivalent to each other, but (6a) is suitable for a perturbation calculation of the 'mainly magnetic', m.e. localized mode, while (6b) is suitable for the calculation of the 'mainly elastic' localized mode.

These equations can be factored according to the irreducible representations of the linear chain symmetry group (C_1)[17].

Then (6a) and (6b) decouple into two determinants each, of ranks two and one respectively. Both first rank determinants correspond to the symmetrical modes (A_{1g}). For the Hamiltonian of equation (1), the magnetic and elastic modes of this symmetry do not interact.

The energies of the localized mixed states are obtained setting the second rank determinants equal to zero.

If ω_m^0 and ω_p^0 are the frequencies of the localized magnon and phonon modes

$$\det\{1 - G_0^+ V^M\}_{\omega=\omega_m^0} = 0$$

and

$$\det\{1 - F_0 V^P\}_{\omega=\omega_p^0} = 0$$

we can write for instance, for ω near ω_m^0:

$$D_M(\omega) = D_M(\omega_m^0) + D'_M(\omega_m^0)(\omega - \omega_m^0)$$

and obtain from here the corrected localized magnon frequency:

$$\omega_m = \omega_m^0 - \frac{D_M(\omega_m^0)}{D'_M(\omega_m^0)}. \qquad (7.1)$$

Since $D_M(\omega_m^0)$ vanishes for $V^{PM} = 0$ the correction term will be small for small V^{PM} if the factors multiplying V^{PM} do not diverge. This will be the case if $\omega_m^0 = \omega_m^0$ since then the 'dressed' phonon Green's function F will be singular at $\omega = \omega_m^0$.

(b) *Scattering states*

Several different cases arise according to the relative width of the magnon and phonon bands, their relative position, and to the nature of the bound or virtual states. For simplicity we assume $H_z = 0$.

We shall discuss here the case where the phonon band is wider than the magnon band. This is the situation in several ferromagnetic insulators where the Heisenberg Hamiltonian is assumed to be a good model for the description of their magnetic properties (EuS[24], EuO[25], CrO_2[26], $CrBr_3$[27], $GdCl_3$, four Cu salts and one of Dy[28]). We will further limit ourselves to the discussion of the effect of the m.e. coupling on the scattering of phonons due to impurities. If the frequency of the incoming phonon is higher than the maximum magnon frequency, the formal solutions of equations (8) (neglecting the excitation of a^*) are:

$$a = (1 - G_0^+ V_{\text{eff}})^{-1} G_0^+ V^{MP} (1 - F_0 V^P)^{-1} x_0 \qquad (8a)$$

and

$$x = x_0 + F(V^P + V^{PM}(1 - G_0^+ V_{\text{eff}})^{-1} G_0^+ V^{MP} \times (1 - F_0 V^P)^{-1}) x_0. \qquad (8b)$$

If for simplicity we assume no variation of the purely elastic parameters at the impurity site, ($V^P = 0$), the origin of phonon scattering lies in the magnetoelastic coupling. With $x_0 = e^{in\phi}$ (see f.e. [18]) we obtain:

$$x_n = e^{in\phi} - \frac{i4(V' - V)}{mh\omega_2^3 \Delta(\epsilon)} \frac{\alpha^2(\epsilon + 1)^3}{(1 - (\alpha/4)^2(\epsilon + 1)^2)^{1/2}} [\Delta_{22} g_0 - \sqrt{2}\Delta_{21} g_1] e^{i|n|\phi} \qquad (9.a)$$

where

$$\omega_2 = \frac{4r}{m}; \quad g_0 = 2JSG_{00};$$

$$G_{ij} = \frac{1}{2JSN} \sum_q \frac{e^{i\bar{q}\cdot(\bar{R}_i - \bar{R}_q)}}{\epsilon - \epsilon_q}$$

$$\text{sen}^2\phi_2 = \frac{\omega_3^2}{\omega_2}; \quad \epsilon = \frac{\omega}{2\omega_1} - 1;$$

$$\alpha = \frac{\omega_1}{\hbar\omega_2} = 4\left(\frac{m}{r}\right)^{1/2} \frac{JS}{\hbar}$$

$\Delta(\epsilon)$ is the determinant of the second rank submatrix corresponding to the A_{1u} modes; Δ_{22}, Δ_{21} are elements of this determinant. If we assume that in the absence of m.e. coupling a localized magnetic mode exists at $\omega_m^0 (2\omega_1 < \omega_m^0 < \omega_2)$, we can now write for small $(V' - V)$ and ω near ω_m^0 (see[21]

$$\Delta_1 \simeq \omega - \omega_m^0 + \frac{i\Gamma}{2}$$

where

$$\omega_m = \omega_m^0 - \frac{RR' + II'}{R'^2 + I'^2} \simeq \omega_m^0 - \frac{II'}{R'^2}$$

is the resonance frequency and

$$\Gamma = \frac{2(IR' - RI')}{R'^2 + I'^2} \simeq \frac{2I}{R'}$$

is the line width. Here R, I are the real and imaginary parts of Δ_1, and R', I' are their derivatives with respect to ω, evaluated at $\omega = \omega_m^0$.

Since Δ_1 appears in the denominator of the expression giving the amplitude $a(\omega)$ ((8.1)) it follows that $a(t)$ will decay exponentially with decay time $1/\Gamma$.

The case of scattering of phonons whose frequency lies inside the magnon band, can be discussed along similar lines. The most important difference with the case in which $V^{PM} = 0$, will arise in the vicinity of the resonant magnon frequencies.

In this case resonant scattering of phonons will also occur, but the width of the resonance will not only be given by V^{MP} but also by the natural width of the magnon mode.

3. CONCLUSIONS

From the previous calculations we conclude that:

(a) Whenever the uncoupled localized or/and virtual states have sufficiently close energies, there will be a substantial modification of their dynamics due to the m.e. interaction. This will affect those observable properties that depend on the dynamics of the localized modes (as in the Mösbauer effect). Furthermore, since the magnon modes have frequencies which are temperature dependent (see [6], [7], [10]), a calculation similar to ours at finite temperatures would be extremely desirable.

(b) The lifetime of the running wave phonon (magnon) modes may be affected by the existence of localized magnon (phonon) modes of neighbouring energies, through the m.e. interaction. Thus those phenomena which depend preponderantly on running wave modes of given frequencies, will be influenced if the latter happen to lie close to the localized states (thermal conductivity).

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