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A simple cubic magnetic semiconductor superlattice in the narrow-band limit

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Abstract. In recent years advances in materials science in the area of molecular beam epitaxy have made possible the synthesis of novel materials and novel structures such as superlattices and multilayers. In the face of the expanding experimental interest in superlattices there is a need for theoretical work to be done. In addition there has been a lot of attention focused on magnetic semiconductors (MS), materials whose magnetic and semiconducting properties interact with each other. In this paper a MS superlattice is investigated using a Green function method applied to the s–d interaction model. The spectrum of spin-wave excitations is calculated and the spectral intensity of certain spin-wave modes is derived from a consideration of the imaginary part of the Green function.

1. Introduction

There has been much recent experimental interest in the study of magnetic semiconductors (MS). Materials which can be classed as ferromagnetic semiconductors include europium chalcogenides (such as EuO and EuS) and spinels (such as $CdCr_2S_4$ $CdCr_2Se_4$ and $HgCr_2Se_4$). In a MS a clear distinction can be made between the localized and the itinerant spins. The localized spins consist of f- or d-shell electrons, giving rise to a lattice of spins. The itinerant spins, on the other hand, are the conduction electrons from the outermost s-shell and they can hop from (atomic) site to site. The interaction of these two types of spin gives rise to the interesting properties of MSs.

Anderson [1] showed that the d electrons in magnetic insulator compounds are localized. The spin-wave theory of ferromagnetic metals was developed by Herring and Kittel [2] and this is the basis for the itinerant part of the present theory. For a discussion of the background to this topic, see for example [3].

Additionally, the development of molecular beam epitaxy, by which crystals can be grown one layer at a time, has paved the way for the investigation of artificially layered materials such as superlattices. A superlattice is a regularly repeating structure made up of set number of molecular layers of each of its components. In this paper a binary superlattice is considered, i.e. two materials A and B which alternate in an infinite laminate. The 'unit cell' has $(n_A - 1)$ layers of material A and $(n_B - 1)$ layers of material B. The material is assumed to be effectively infinite in the plane of these layers.

The theory of excitations in ferro- and antiferromagnetic superlattices has been developed [4]; in this case the superlattice consisted of alternate slabs of magnetic and non-magnetic material. Also, a calculation for a ferromagnetic multilayer in the dipolar–magnetostatic limit [8] has been done. Light-scattering theory for a stack of ferromagnetic

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slabs (a magnetic/non-magnetic superlattice) has also been developed [9]. There is much experimental interest in magnetic superlattices (e.g. [10, 11]).

In this paper a microscopic approach [14] to the study of excitations in a magnetic semiconductor superlattice is used in which a Heisenberg ferromagnet interacts with the itinerant spins of conduction electrons via a contact-type interaction. A calculation [12] based on the same microscopic approach has been performed for an *infinitely extended* bulk magnetic semiconductor and it shows that there are two spin-wave branches ('acoustic' and 'optical') and a continuum of Stoner-like magnetic excitations.

The same model has been examined in some recent papers [17, 18] for the cases of a semi-infinite material, a finite slab and an interface between two dissimilar semi-infinite media. Building on those works, this paper considers the case of an infinite-bulk binary superlattice with a simple cubic unit cell.

In a superlattice the excitations can be divided into three types [13]: 'pure bulk' (which propagate throughout the whole material and have bulk-like character in both component materials), 'pure interface' (which are exponentially decaying in both materials and therefore confined to the interfaces) and 'bulk–interface' (which have bulk character in one material and decay exponentially in the other). This classification is discussed further in section 3.

The parameters used in this paper for material A correspond to the case of $CdCr_2Se_4$ [5], which is usually considered to be a narrow-band magnetic semiconductor. Then the parameters for material B are varied in turn to give the figures presented here.

Experimental observation of spectra in these materials has been achieved via light scattering [6]. The acoustic branch modes shown in this paper are very accessible to Brillouin scattering techniques.

The purpose of this paper is to investigate the excitation spectra for a MS superlattice, using a Green function approach

2. The Green function theory

Beginning with the Hamiltonian

$$\mathcal{H} = \frac{1}{2}\sum_{i,j,\sigma} t_{ij} a^{\dagger}_{i\sigma} a_{j\sigma} - \frac{1}{2}\sum_{i,j} J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - \sum_i I_i \boldsymbol{S}_i \cdot \boldsymbol{s}_i$$
$$- g\mu_B H_0 \sum_i S^z_i - g_e \mu_B H_0 \sum_i s^z_i \qquad (1)$$

the narrow-band limit is considered, that is where the conduction bandwidth (which is equal to $12t$ for a simple cubic lattice) is small compared to IS. Consequently, the 'hopping' interaction strength t is set to zero in the remainder of this paper.

In the above equation the operators $a^{\dagger}_{i\sigma}$ and $a_{i\sigma}$ are creation and annihilation operators respectively for a conduction electron at site i and having spin $\sigma = \pm 1$. The spin operators for conduction electrons are defined in terms of the a and $a^{\dagger}$; $s^+ = a^{\dagger}_{i+} a_{i-}$, $s^- = a^{\dagger}_{i-} a_{i+}$, $s^z = \frac{1}{2}(a^{\dagger}_{i+} a_{i+} - a^{\dagger}_{i-} a_{i-})$. The vector operators $\boldsymbol{S}_i$ are spin operators for the localized d- or f-shell electrons. Thus the term in J_{ij} is a Heisenberg-like interaction between nearest-neighbour sites on the lattice. H_0 is the external magnetic field, g and g_e are the Landé factors for localized and conduction electrons respectively and μ_B is the usual Bohr magneton.

The superlattice structure consists of $(n_A - 1)$ layers of material A and $(n_B - 1)$ layers of material B infinitely repeated in the z-direction and the material is assumed to be infinite in the (x, y)-plane. The interaction J_{ij} is assumed to take a value J_A in the bulk of material

A and J_B in the bulk of material B. For nearest neighbours in the the layer in material A adjoining the interface the value is taken as J_{AS} and similarly in material B the value is J_{BS}. When one site is in material A and one in B (i.e. across the interface) the value taken is J_I. For I_i the situation is similar except for the fact that it is a single-site interaction, so there is no I_I, but all the other modifications apply.

The Green function can be derived via an equation of motion:

$$\omega\langle\langle S_r^+; S_{r'}^-\rangle\rangle = \frac{1}{2\pi}\langle[S_r^+, S_{r'}^-]\rangle + \langle\langle [S_r^+, \mathcal{H}]; S_{r'}^-\rangle\rangle. \tag{2}$$

The Green functions appearing in this equation have been Fourier transformed in time and are the frequency- (ω-) dependent versions; the subscript to indicate this explicitly has been dropped.

After calculating the commutators in (2) I arrive at an equation involving terms like $\langle\langle S_r^z S_r^+; S_{r'}^-\rangle\rangle$. In principle one could derive a further equation for these Green functions, which would then involve terms in yet more complicated Green functions. However, in order to perform some calculations it is necessary to truncate this hierarchy of Green functions and the approximation chosen to do this is the RPA or random-phase approximation: this replaces the operator S^z by its average value $\langle S^z\rangle \simeq S$ and is valid in a low-temperature regime, where the spins do not vary much from the fully aligned state.

Having done this it is possible to proceed towards a solution of the equations. Taking a Fourier transform in the (x, y)-plane using the lattice periodicity (lattice spacing $= a$) and in the z-direction using the superlattice periodicity ($(n_A-1)a+(n_B-1)a = (N-2)a = L$), the following expression is obtained:

$$\frac{S_m}{\pi}\delta_{m,n} = \left(\omega - \omega_0 + I_m\langle s_m^z\rangle + 4J_m S_m(1-\gamma(k_\parallel)) + J_{m,m-1}S_{m-1} + J_{m,m+1}S_{m+1}\right)G_{m,n}$$
$$-J_{m,m+1}S_m G_{m+1,n} - J_{m,m-1}S_m G_{m-1,n} - I_m S_m g_{m,n} \tag{3}$$

where $\omega_0 = g\mu_B H_0$, $G_{m,n}(k_\parallel, \omega)$ is the Fourier transform of the Green function $\langle\langle S_r^+; S_{r'}^-\rangle\rangle$ and similarly $g_{m,n}(k_\parallel, \omega)$ is the Fourier transform of $\langle\langle s_r^+; S_{r'}^-\rangle\rangle$. The function $\gamma(k_\parallel) = \frac{1}{2}(\cos(k_x a) + \cos(k_y a))$ arises from the 2D Fourier transform and the a in the argument of the cosine functions is the simple cubic lattice spacing.

A similar equation to (3) holds for the $g_{m,n}$ but in this case, since I am assuming $t_{i,j} \equiv 0$, the corresponding equation is much simpler. It reduces to

$$g_{m,n} = \frac{I_m\langle s_m^z\rangle}{\omega - \omega_e - I_m S_m}G_{m,n} \tag{4}$$

where $\omega_e = g_e\mu_B H_0$. By combining (3) and (4) an equation for $G_{m,n}$ can be derived and it is found [16] that a convenient representation is in terms of tridiagonal matrices, whereby this equation can be solved. The resulting equation is:

$$(M^0_{ml} + \Delta_{ml})G_{ln} = \frac{S_m}{\pi}\delta_{m,n} \qquad 0 < m, n \leqslant N-2 \tag{5}$$

where $\mathbf{M}^0$ is the pure tridiagonal matrix and Δ_{ml} includes the effect of the interfaces:

$$M^0_{lm} = \begin{cases} J_A S_A(a_A\delta_{m,l} - \delta_{m,l\pm1}) & \text{for } 0 < l,\ m \leqslant n_A - 1 \\ J_B S_B(a_B\delta_{m,l} - \delta_{m,l\pm1}) & \text{for } n_A - 1 \leqslant l,\ m \leqslant N-2 \end{cases} \tag{6}$$

where a_A, a_B and Δ_{lm} are given by

$$a_A J_A S_A = \omega_0 - \omega + 2J_A S_A(3 - 2\gamma(k_\parallel)) + I_A\langle s_A^z\rangle + \frac{I_A^2\langle s_A^z\rangle}{\omega - \omega_e - I_A S_A} \tag{7}$$

with a similar expression for a_B but with the As being replaced by Bs. The interface perturbation Δ_{lm} is given by the following equation:

$$\begin{aligned}
&\Delta_{1,1} = \Delta_{n_A-1,n_A-1} = J_A S_A \Delta_A && \Delta_{n_A,n_A} = \Delta_{N-2,N-2} = J_B S_B \Delta_B \\
&\Delta_{n_A-1,n_A} = -J_I S_A && \Delta_{n_A,n_A-1} = -J_I S_B \\
&\Delta_{N-2,1} = -J_I S_B \exp\{ik_z L\} && \Delta_{1,N-2} = -J_I S_A \exp\{-ik_z L\} \\
&\Delta_{l,m} = 0 && \text{otherwise.}
\end{aligned} \tag{8}$$

The quantity Δ_A (and in a similar manner, Δ_B, but with B replacing A and vice versa) is defined as

$$\Delta_A = -1 - 4(1-\gamma(k_\parallel))(1 - J_{AS}/J_A) - \frac{I_A\langle s_A^z\rangle}{J_A S_A}\left(1 - \frac{I_{AS}\langle s_{AS}^z\rangle}{I_A\langle s_A^z\rangle}\right) - \frac{J_I S_B}{J_A S_A} - \frac{I_A^2\langle s_A^z\rangle}{J_A(\omega - \omega_e - I_A S_A)} + \frac{I_{AS}^2\langle s_{AS}^z\rangle}{J_A(\omega - \omega_e - I_{AS} S_A)} \tag{9}$$

where J_{AS} represents the modified interaction in the set of layers on one side of the interfaces. Similarly, I_{AS} and $\langle s_{AS}^z\rangle$ are the modified values of I_A and $\langle s_A^z\rangle$ in that same set of layers. The solution for the Green function is derived by inverting the tridiagonal matrix of elements M_{lm}^0. The matrix splits into two blocks each of which can be inverted by a standard well-known formula [19] by defining $x + x^{-1} = a_A$ and $y + y^{-1} = a_B$. Then the inverse is

$$(M^0)_{lm}^{-1} = \begin{cases} \dfrac{x^{l+m} - x^{|l-m|} + x^{2n_A-l-m} - x^{2n_A-|l-m|}}{J_A S_A (x - x^{-1})(1 - x^{2n_A})} & 1 \leqslant l, m \leqslant n_A - 1 \\ \dfrac{y^{2+l+m-2n_A} - y^{|l-m|} + y^{N-2-l-m} - y^{2n_B-|l-m|}}{J_B S_B (y - y^{-1})(1 - y^{2n_B})} & n_A \leqslant l, m \leqslant N-2 \\ 0 & \text{otherwise.} \end{cases} \tag{10}$$

The formal solution for the Green function is therefore

$$G_{m,n}(k_\parallel, \omega) = \left[\mathbf{I} + (\mathbf{M}^0)^{-1}\Delta\right]^{-1} (\mathbf{M}^0)^{-1} \frac{S_n}{\pi}. \tag{11}$$

3. The excitation spectrum

The poles of the Green function give the energy excitation spectra of the material. To calculate the poles it is sufficient to examine the determinant of the quantity in square brackets in (11). Call this determinant D_0. Then look for solutions of $D_0 = 0$. Calculating the determinant and writing $X_1 = (x - x^{(2n_A-1)})/(1 - x^{(2n_A)})$, $X_2 = (x^2 - x^{(2n_A-2)})/(1 - x^{(2n_A)})$ and $X_3 = (x^{(n_A-1)} - x^{(n_A+1)})/(1 - x^{(2n_A)})$ and with similar expressions for Y_1, Y_2 and Y_3 (involving y and n_B) the following equation is derived:

$$\begin{aligned} D_0 = {} & (1 + 2\Delta_A X_1 + \Delta_A{}^2 X_2)(1 + 2\Delta_B Y_1 + \Delta_B{}^2 Y_2) \\ & + 2r_I[(X_1 + \Delta_A X_2)(Y_1 + \Delta_B Y_2) + \cos(k_z L) X_3 Y_3] + r_I^2 X_2 Y_2 \end{aligned} \tag{12}$$

where $r_I = J_I^2/(J_A J_B)$. Note that when the interaction strength across the interface vanishes, so that $r_I = 0$ then the above equation simplifies to

$$0 = (1 + 2\Delta_A X_1 + \Delta_A{}^2 X_2)(1 + 2\Delta_B Y_1 + \Delta_B{}^2 Y_2) \tag{13}$$

which is simply the product of the two separate equations for a finite slab of material A and one of material B. This is what would be expected after all, since when $r_I = 0$ there is

no connection across the interfaces and the problem reduces to two infinite sets of identical slabs.

As with other cases of simpler geometry the nature of the modes depends on the variables x and y. In general a mode will be localized in material A if $|x| < 1$ and a mode will be 'bulk-like' in material A if $|x| = 1$. Similar conditions on y apply for material B. Taken together these conditions on x and y imply the classification [13]. However, the modes observed in the figures in this paper connect smoothly across the bulk-band boundaries and so one branch may have differing properties along its length in $k_\parallel$ space.

The excitation spectrum for infinitely extended bulk material A (and with a similar formula for material B) is given by the following equation:

$$\omega - \omega_0 - 2S_A J_A \left[3 - 2\gamma(k_\parallel) - \cos(q_z a)\right] - I_A\langle s_A^z\rangle - \frac{I_A^2 S_A \langle s_A^z\rangle}{\omega - \omega_e - I_A S_A} = 0. \tag{14}$$

In figures 1–5, which show the excitation spectra, the limits of the bulk bands are shown by heavier lines. These lines correspond to taking $\cos(q_z a) = \pm 1$ in (14). There are two regions of energy, the 'acoustic' $\omega \ll I/J$ and the 'optic' $\omega \sim I/J$; in this paper only the 'acoustic' region is considered for simplicity and also because the acoustic region is easier to observe experimentally than the optic modes. Mauger and Mills [7] found that within RPA the acoustic mode is not damped and their treatment beyond the RPA finds that the damping of the acoustic modes only becomes large as $T \to T_C$. Therefore, the acoustic mode results displayed here should be observable, at least for temperatures low compared with the Curie temperature.

To simplify matters further I shall assume that $I_{AS} = I_A$, $I_{BS} = I_B$, $\langle s_{AS}^z\rangle = \langle s_A^z\rangle$, $\langle s_{BS}^z\rangle = \langle s_B^z\rangle$. The figures are all plotted with 'in-plane wavevector' as their x-axis; in fact, it is k_x which is used as the independent variable with k_y set to zero.

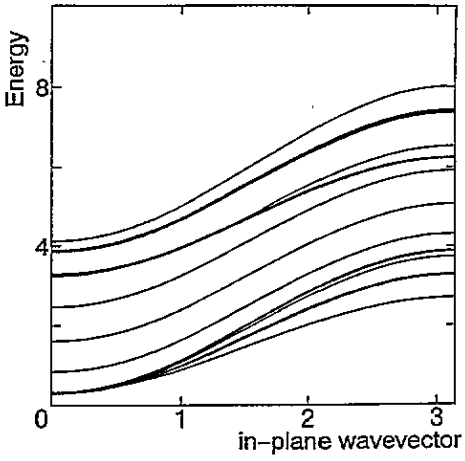

Figure 1. The excitation spectrum in the acoustic region: heavy lines show the limits of the bulk bands, parameter values are $J_A = 1.0$, $J_B = 1.2$, $J_{AS} = 0.5$, $J_{BS} = 0.8$, $J_I = 2.0$, $I_A = I_{AS} = 200$, $I_B = I_{BS} = 240$, $\langle s_A^z\rangle = \langle s_{AS}^z\rangle = \frac{1}{2}$, $\langle s_B^z\rangle = \langle s_{BS}^z\rangle = \frac{1}{2}$, $S_A = S_B = \frac{3}{2}$, $\omega_0 = \omega_e = 0.3$, $k_z L = 0$, $n_A = 3$, $n_B = 7$.

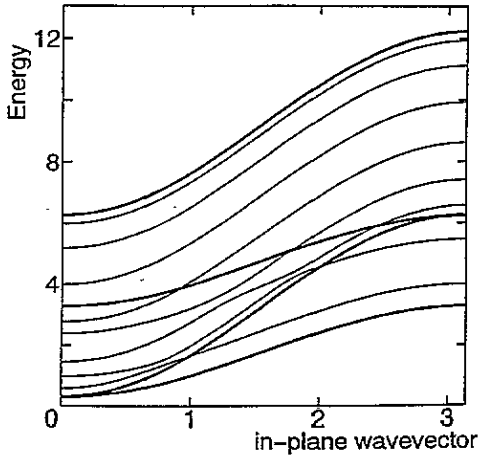

Figure 2. The excitation spectrum in the acoustic region with $S_A = \frac{3}{2}$ and $S_B = \frac{5}{2}$ and other parameters as for figure 1.

Figure 1 shows a general case of discrete bulk-like modes and two interface modes labelled. Figure 2 shows the mode mixing which can occur when the parameters of the two materials are sufficiently different; here the magnitudes of the localized spins are $S_A = \frac{3}{2}$ and $S_B = \frac{5}{2}$. Figure 3 shows the effect of having a small value of J_I; note how the modes follow one or other of the bulk-band limits; in this case it is very much like having two separate materials. Figure 4 shows the result of altering $\cos(k_z L)$ from the parameters of figure 1; note the difference in the uppermost bulk mode in this figure. Figure 5 shows the effect of altering the thickness of one of the components of the superlattice (here $n_A = 5$). The two interface modes in this figure have been labelled p and q for comparison with figure 6, which is discussed in section 4. Note how the lower interface mode (labelled q) is lowered from the case of figure 1. This lowering reaches a minimum when the two components have equal thickness as would be expected intuitively. The dependence on relative thickness of the component layers has not been investigated fully and is a possibility for further work; for example, does the ratio $J_A S_A / J_B S_B$ affect this effect in any way?

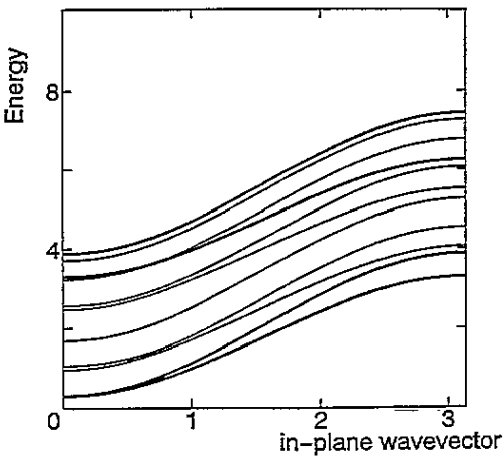

Figure 3. The excitation spectrum in the acoustic region with $J_I = 0.1$ and other parameters as for figure 1.

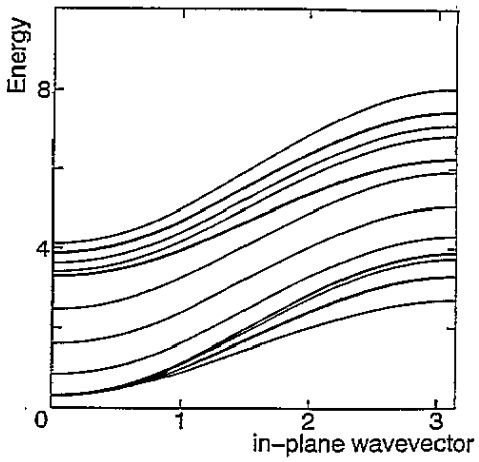

Figure 4. The excitation spectrum in the acoustic region with $k_z L = \pi$ and other parameters as for figure 1.

4. Spectral intensity

As an example of the application of the Green function method, the explicit Green function is caculated for a particular case and from that result the spectral intensity, which is related to the imaginary part of the Green function, is calculated. Thus the explicit form of the Green function in the topmost layer of material 'A' is given by the following:

$$G_{1,1}(k_\parallel, \omega) = \frac{(X_1 + \Delta_A X_2)(1 + 2\Delta_B Y_1 + \Delta_B^2 Y_2) - r_1 X_2 (Y_1 + \Delta_B Y_2)}{J_A \pi D_0}. \tag{15}$$

The imaginary part of the Green function is related to the spectral intensity; there is also a thermal factor, but if low temperature is assumed this can be effectively set to unity. The imaginary part of the above expression will have delta-function contributions at the

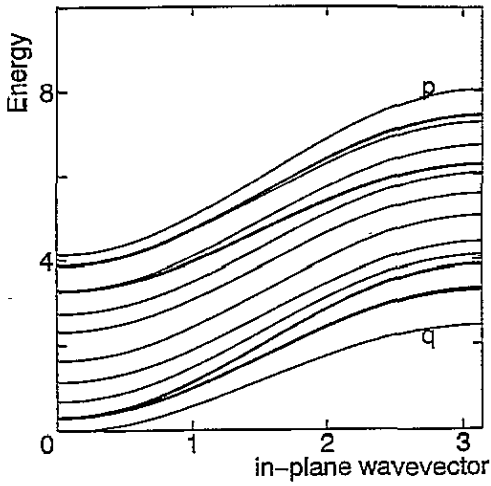

Figure 5. The excitation spectrum in the acoustic region with $n_A = 5$ and other parameters as for figure 1. Two interface modes are marked p and q for comparison with figure 6.

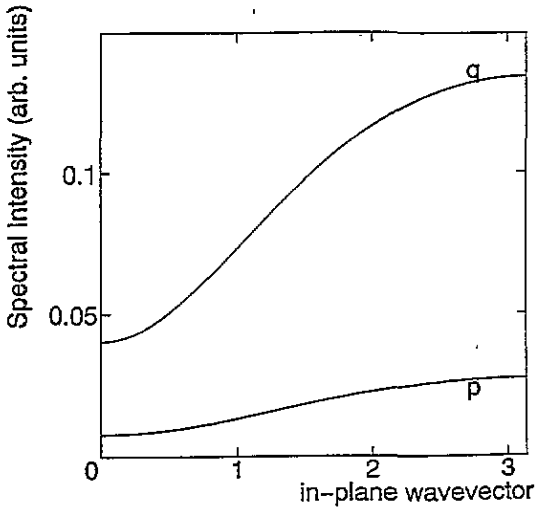

Figure 6. The spectral intensity in layer 1 (top layer of material A slab) for the interface modes in figure 5, correspondingly labelled p and q.

energies corresponding to the interface modes (x and y are both real). To calculate the contributions to the spectral intensity from the two interface modes labelled p and q in figure 5 the following formula is employed:

$$\text{If } F(x) = \frac{f(x)}{g(x)} \qquad \text{then } \Im(F(x)) = \sum_{x_r} \frac{f(x)}{g'(x)} \delta(x - x_r) \tag{16}$$

where x_r is a root of $g(x) = 0$.

Thus, the derivative $\mathrm{d}D_0/\mathrm{d}\omega$ is calculated and used to compute the imaginary part of the Green function. The results are shown in figure 6 where the labels p and q correspond to the two interface modes of figure 5.

5. Conclusion

The Green function for a magnetic semiconductor binary superlattice having simple cubic crystal structure has been calculated and the result has been used to determine excitation spectra. The effects of varying the parameters of the model, such as the interaction strength across the interfaces and the magnitude of the localized spins, has been investigated and the resulting graphs have been presented. Also the spectral intensity of the interface modes in the top layer of material A has been calculated.

The model considered here has been limited in many ways, by assuming $t \equiv 0$ or by not considering $I_{AS} \neq I_A$ for instance, and it would be interesting to investigate some extensions of the theory presented here in future. Also, in order to facilitate the application of this work to experimental investigations a calculation of, for example, the spin-wave stiffness parameter could be performed. There has not been time to include such a calculation here, but I hope to consider this in future work.

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