Linear spin-wave theory of incommensurably modulated magnets

Timothy Ziman*
Institut Laue-Langevin, 156X, 38042 Grenoble Cédex, France

Per-Anker Lindgård
Risø National Laboratory, DK-4000 Roskilde, Denmark
(Received 22 May 1985)

Calculations of linearized theories of spin dynamics encounter difficulties when applied to incommensurable magnetic phases: Lack of translational invariance leads to an infinite coupled system of equations. We resolve this for the case of a "single-Q" structure by mapping onto the problem of diagonalizing a quasiperiodic Hamiltonian of tight-binding type in one dimension. This allows for calculation of the correlation functions relevant to neutron scattering or magnetic resonance experiments. With the application to the case of a longitudinally modulated magnet a number of new predictions are made: at higher frequency there appear bands of response sharply defined in frequency, but broad in momentum transfer; at low frequencies there is a response maximum at the q vector corresponding to the modulation vector. We discuss generalizations necessary for application to rare-earth magnets.

INTRODUCTION

The increasing availability of large single crystals of rare-earth metals and their compounds has revived experimental attention in the dynamics of their magnetically ordered phases, in particular the sinusoidally modulated phases of neodymium, praseodymium, and $CeAl_2$.[1] There is, however, a difficulty in the interpretation of inelastic experiments, in that linear spin-wave theory, the normal starting point for understanding excitations in magnets, is not straightforward when the average moment varies from site to site. This was recognized long ago.[2] The difficulty is precisely what makes the materials interesting in a wider theoretical context: In the presence of incommensurable modulation the magnetic structure lacks translational invariance and therefore crystal momentum $\mathbf{q}$ is not a good quantum number. The same is true for helical phases if we include effects of anisotropy-induced bunching, as has been observed in the helical phase of holmium.[3] The linear spin-wave equations, which for a commensurable structure may be solved by diagonalizing a finite-dimensional matrix, form an infinite coupled set. There have been attempts to cope with this by truncation or by perturbation theory:[4] such efforts have had limited success but the approximations made are not well controlled, and in some cases have led to misleading results.[5]

Fortunately, there has been a body of work on the mathematical physics of several closely related problems involving quasiperiodicity, inspired by the dynamics of structurally modulated systems and charge-density waves[6] and by theories of electronic motion in a film in the presence of a transverse magnetic field.[7] Consequently, for the case of a "single-Q" structure, to which this paper will be restricted, the properties of the coupled equations are understood, and we can calculate the inelastic spin-wave cross section in a single-magnon approximation. While we limit this discussion to a simplified model Hamiltonian, the method can be generalized in a straightforward manner to incorporate a fuller description of the anisotropies. A number of features appear that previously were unexpected and should be observable in experiments. The structure predicted by the present, nonperturbative account is a good deal richer than that of broadened magnons found by previous treatments.[4]

LINEAR SPIN-WAVE THEORY

For simplicity, we consider an exchange Hamiltonian that is quadratic in the spin operators (later we shall outline the extension to higher-order anisotropies),

$$\mathscr{H}=-\int_{\mathrm{BZ}}\left(J(\mathbf{q})\tfrac{1}{2}(S_{\mathbf{q}}^{+}S_{\mathbf{q}}^{-}+S_{\mathbf{q}}^{-}S_{\mathbf{q}}^{+})-K(\mathbf{q})S_{\mathbf{q}}^{z}S_{\mathbf{q}}^{z}\right)d^{d}q\ , \tag{1}$$

where the integral is over the Brillouin zone (BZ). To be more specific, we take the simplified model introduced by Elliott[8] for rare earths, in which we imagine competing exchange interactions J_1 and J_2 between nearest and next-nearest neighbors along a single crystalline axis $\mathbf{a}$, a transverse ferromagnetic exchange J_0, and a strong uniaxial anisotropy favoring a single spin axis $\mathbf{z}$,

$$J(\mathbf{q})=J_1\cos(\mathbf{q}\cdot\mathbf{a})+J_2\cos(2\mathbf{q}\cdot\mathbf{a})+J_0[\cos(\mathbf{q}\cdot\mathbf{b})+\cos(\mathbf{q}\cdot\mathbf{c})]\ ,$$

$$K(\mathbf{q})=D+J(\mathbf{q})\ ,$$

$$S_{\mathbf{q}}^{\alpha}=\sum_{r}e^{i\mathbf{q}\cdot\mathbf{r}}S_{r}^{\alpha}\ .$$

In a random-phase approximation, if $J(\mathbf{q})$ has a maximum at $\mathbf{q}=\mathbf{Q}=Q\mathbf{a}$ such that $\cos Q=-J_1/4J_2$, there is

a stable ground state satisfying

$$\langle S_{\mathbf{r}}^z \rangle = S\cos(\mathbf{Q}\cdot\mathbf{r}) ,$$
$$\langle S_{\mathbf{q}}^z \rangle = \frac{S}{2}[\delta(\mathbf{q}-\mathbf{Q})+\delta(\mathbf{q}+\mathbf{Q})] , \tag{2}$$

provided D is sufficiently strong to ensure that this longitudinally modulated phase is of lower energy than a helical phase,[9]

$$D \geq J(\mathbf{Q}) .$$

Note that the ground state has a broken phase symmetry, i.e., there is an arbitrary phase ϕ in the spin density (2) that we have taken to be equal to zero. In principle, a small field conjugate to this variable should be taken which is set to zero at the end of the calculation. To the level of the linearized theory presented here, following Ref. 2, this will not affect the final equations; however, this broken symmetry has consequences which we shall mention below.

Using the equation of motion for spin operators, in units such that $\hbar=1$,

$$i\frac{dS_{\mathbf{q}}^+}{dt} = [S_{\mathbf{q}}^+, \mathcal{H}] = \sum_{\mathbf{q}'} K(\mathbf{q}')S_{\mathbf{q}+\mathbf{q}'}^+ S_{\mathbf{q}'}^z - J(\mathbf{q}')S_{\mathbf{q}'}^+ S_{\mathbf{q}-\mathbf{q}'}^z . \tag{3}$$

Within the linearized approach we replace operators S^z in (3) by their expectation values (2),

$$i\frac{dS_{\mathbf{q}}^+}{dt} = \frac{S}{2}[K(\mathbf{Q})-J(\mathbf{q}+\mathbf{Q})]S_{\mathbf{q}+\mathbf{Q}}^+ + \frac{S}{2}[K(\mathbf{Q})-J(\mathbf{q}-\mathbf{Q})]S_{\mathbf{q}-\mathbf{Q}}^+ . \tag{4}$$

Making the substitution $\mathbf{q}\leftrightarrow\mathbf{q}+n\mathbf{Q}$, we derive the infinite set of coupled operator equations,

$$i\frac{d}{dt}S_{\mathbf{q}+n\mathbf{Q}}^+ = W_{n+1}S_{\mathbf{q}+(n+1)\mathbf{Q}}^+ + W_{n-1}S_{\mathbf{q}+(n-1)\mathbf{Q}}^+ ,$$
$$W_n = \frac{S}{2}[K(\mathbf{Q})-J(\mathbf{q}+n\mathbf{Q})] . \tag{5}$$

Thus within a linearized theory, for $\mathbf{Q}$ incommensurable with the lattice, the normal modes and their frequencies are derived from an infinite-dimensional matrix with first off-diagonal elements only. The hierarchy (5) is as found by Liu and Lindgård:[4] Henceforth our analysis will differ. By the assumption of a single-$\mathbf{Q}$ structure (2), we note that the set (5) has a one-dimensional structure. As (5) stands, however, the matrix is not symmetric: Nonetheless, it may be made so if we define transformed operators

$$a_n^+ = (W_n)^{1/2}S_{\mathbf{q}+n\mathbf{Q}}^+ . \tag{6}$$

Note that D is positive in order to stabilize the longitudinal modulated phase; thus the W_n are positive.

Then the equations of motion for operators a_n^+ are those of a one-dimensional tight-binding model,

$$\mathcal{H} = \sum_n t_n(a_{n+1}^+ a_n + a_n^+ a_{n+1}) , \tag{7}$$

with quasiperiodic coefficients $t_n=(W_nW_{n+1})^{1/2}$. Note that "site n" refers to the operator $S_{\mathbf{q}+n\mathbf{Q}}^+$. If (7) can be diagonalized, the dynamical susceptibility

$$\chi^{+-}(\mathbf{q}',\omega) = i\int_0^\infty e^{i\omega t}\langle [S_{\mathbf{q}'}^+(t), S_{\mathbf{q}'}^-(0)]\rangle dt$$

for spin components transverse to the direction of spin ordering z is determined for the momenta $\mathbf{q}'$ defined by adding integral multiples of the modulation vector $\mathbf{Q}$ to the starting vector $\mathbf{q}$. The angular brackets $\langle\ \rangle$ denote a quantum-mechanical expectation value in the ground state.

Expanding operators $S_{\mathbf{q}'}^+$ in terms of the eigenmodes and using the boundary condition (2), we find

$$\mathrm{Im}\chi^{+-}(\mathbf{q}',\omega) = \sum_\alpha \omega_\alpha \left| \sum_{n(\mathbf{q}',\mathbf{q},\omega)} \frac{f_\alpha(n)}{(W_n)^{1/2}} \right|^2 \delta(\omega-\omega_\alpha) , \tag{8}$$

where $n(\mathbf{q}',\mathbf{q},\omega)$ denotes the set of integers such that $\mathbf{q}'$ and $\mathbf{q}+n\mathbf{Q}$ coincide when reduced to the first Brillouin zone, and $b_\alpha^+ = \sum_{-\infty}^{\infty} f_\alpha(n)a_n^+$ is the eigenmode of energy ω_α. If $\mathbf{Q}$ is incommensurable, $n(\mathbf{q}',\mathbf{q},\mathbf{Q})$ has at most one element, and

$$\mathrm{Im}\chi^{+-}(\mathbf{q}+n\mathbf{Q},\omega) = \sum_\alpha \frac{\omega_\alpha |f_\alpha(n)|^2}{W_n}\delta(\omega-\omega_\alpha) . \tag{9}$$

The set $\{\mathbf{q}+n\mathbf{Q}\}$, reduced to the first Brillouin zone, is a dense set of vectors on the line through $\mathbf{q}$ and parallel to $\mathbf{Q}$.

It is apparent from (9) that χ^{+-}/ω is the local density of sites, "local" in the space of sites $\mathbf{q}+n\mathbf{Q}$, and thus formula (9) should converge, in the limit of an infinite crystal, to a measure which corresponds to the nature of the spectrum of the operator: a smooth function of q if the spectrum is continuous, or an infinite set of δ functions, i.e., "satellites," if the spectrum is pure point.

It is instructive to recover the results for a simple *commensurable* magnetic phase: the two-sublattice antiferromagnet with $\mathbf{Q}=\pi\mathbf{a}$. In this case the t_n are equal for any n,

$$t_n = t_m = t(\mathbf{q}) = \frac{S}{2}\sqrt{K(\pi)-J(\mathbf{q})}\sqrt{K(\pi)-J(\mathbf{q}+\pi)} . \tag{10}$$

By translational invariance the eigenoperators are

$$b_k^+ = \sum_n e^{ikn}a_n^+ , \tag{11}$$

with $\omega_k = 2t(\mathbf{q})\cos k$. In this case, however, out of the infinite set of frequencies ω_k, only two are physically relevant, since for all n the operator a_n^+ is identical to a_{n+2}^+. Thus, except when k is 0 or π the factors e^{ikn} interfere destructively, and the operator b_k^+ is the zero operator. We recover, then, for $\mathbf{Q}=\pi\mathbf{a}$ the result that there is a pair of frequencies

$$\omega = \pm 2t(\mathbf{q}) . \tag{12}$$

Note that the wave vectors k are in the reciprocal lattice to the chain of "sites" (and should not be confused with crystal momentum $\mathbf{q}$).

Returning to the *incommensurable* case, the tight-

binding Hamiltonian is aperiodic with the nth off-diagonal element

$$t_n=\frac{S}{2}K(\mathbf{Q})\{1+\alpha\cos(q_a+nQ)+\beta\cos[2(q_a+nQ)]\}^{1/2}$$
$$\times(1+\alpha\cos[q_a+(n+1)Q]$$
$$+\beta\cos\{2[q_a+(n+1)Q]\})^{1/2}, \tag{13}$$

where $\alpha=-J_1/K(\mathbf{Q})$, $\beta=-J_2/K(\mathbf{Q})$, and $q_a=\mathbf{q}\cdot\mathbf{a}$. The Hamiltonian (7) differs from Harper's equation, which is the lattice equivalent of Mathieu's equation, and which has been analyzed by Aubry and others,[10] in that it has no site-diagonal term. In our case it is the off-diagonal term that is modulated. The same methods used, essentially adapted from those applied to random systems, can be applied here. In particular, we can define a complex function $k(E)$, the imaginary part of which is the Lyapunov exponent, the real part the integrated density of states. Simply by multiplying matrices we can rapidly calculate a density of eigenstates and see whether states are extended or localized (on the lattice of sites n).[11] The difference equation for eigenvectors of (7),

$$Ea_n=t_na_{n+1}+t_{n-1}a_{n-1}, \tag{14}$$

rewritten as

$$a_{n+1}=(E/t_n)a_n-\frac{t_{n-1}}{t_n}a_{n-1},$$

is iterated starting from arbitrary boundary conditions (a_0,a_1).

The Lyapunov exponent is defined numerically by

$$\lambda(E)=\lim_{N\to\infty}\left[\frac{\ln||a_N||}{N}\right],$$

and the integral of the density of states

$$I(E)=\int_{-\infty}^{E}\rho(E')dE'=\lim_{N\to\infty}\frac{\Sigma(E,N)}{N}$$

by counting the number of sign changes $\Sigma(E,N)$ of the sequence of real numbers $(a_0,a_1,\ldots,a_N)$. Note that this procedure does not involve diagonalization of the matrix, but gives only spectral information, i.e., it determines $\chi^{+-}(\mathbf{q},\omega)$ averaged over $\mathbf{q}$. In order to calculate eigenvectors and thereby the structure of $\chi^{+-}(\mathbf{q},\omega)$ as a function of $\mathbf{q}$, however, we need to diagonalize a tridiagonal matrix.

SEMICLASSICAL DISCUSSION

The essential properties of the Hamiltonian (7) can be understood within a semiclassical picture, as has been used by Harper[7] and Wilkinson[10] for the case of a Hamiltonian with a diagonally modulated term. Introducing the continuous variable x by

$$\mathbf{q}+n\mathbf{Q}=\mathbf{q}+x\frac{\mathbf{Q}}{(|Q|)}$$

and treating the a_n in (14) as functions of the continuous variable x, we note that

$$a(x+Q)=e^{iQ\hat{k}}a(x) \quad \text{where } \hat{k}=-i\frac{d}{dx}.$$

Equation (14) can therefore be written in operator language,

$$\hat{H}a(x)=Ea(x),$$

where

$$\hat{H}(\hat{x},\hat{k})=t_n(\hat{x})e^{iQ\hat{k}}+e^{-iQ\hat{k}}t_n(\hat{x}). \tag{15}$$

A classical Hamiltonian equivalent to (15) is therefore

$$H(x,k)=2(\cos k)t_n(x)$$
$$=\frac{2SK(\mathbf{Q})}{2}(\cos k)[1+\alpha\cos x+\beta\cos(2x)]^{1/2}$$
$$\times\{1+\alpha\cos(x+Q)+\beta\cos[2(x+Q)]\}^{1/2}. \tag{16}$$

The semiclassical approximation should be valid in the limit $\mathbf{Q}\to\mathbf{0}$ and wave functions with $k\gg Q$, i.e., for a potential varying slowly on the scale of lattice spacing and k. In fact, it appears to explain qualitatively numerical results where these conditions are but barely fulfilled.

To construct a semiclassical wave function we first consider energy contours of the function $H(x,k)$. In Fig. 1 we draw these for k in the range $[0,2\pi]$ and x in the range $[0,4\pi]$. Q is chosen to be small, i.e., the phase is nearly ferromagnetic. α is taken to be -0.386; β is fixed by the choice of Q. These parameters were chosen as a simplified model of neodymium. For larger α there may be further extrema of H, giving greater complexity to the contour diagram. The classical equations of motion have orbits that are either closed, or open in the x direction.

The closed orbits occur around minima A,A' of the surface H or maxima. The contour B separates the closed from the extended orbits whose energies are smaller in absolute value. To lowest order eigenfunctions correspond-

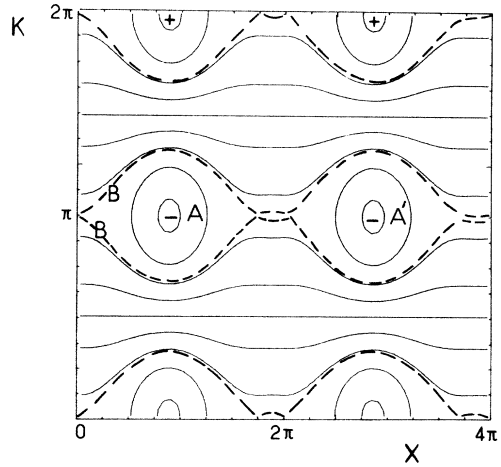

FIG. 1. Energy contours of the classical Hamiltonian (16) with parameters $\alpha=-0.386$, $Q=0.8160$, and $\beta=0.1410$. A and A' are equivalent minima; B is the separatrix separating closed and open orbits. The straight lines $K=\pi/2$ and $3\pi/2$ are contours of energy 0.

ing to a finite set of closed orbits occur at discrete energy levels, in essence those given by Bohr-Sommerfeld quantization. Within a WKB approximation these levels are broadened into a band, the width of which depends on overlap of wave functions localized close to equivalent closed orbits, around A and A', for example. The width is therefore exponentially small for Q approaching a commensurable value. If there is more than one such sharp level, corresponding to the quantization of more than one closed orbit, the overlap, and hence the bandwidth, is larger for those orbits of lower frequency since the states are less tightly bound. Since there are always orbits open in the x direction, we might expect, by comparison to the diagonal problem, that all eigenstates are extended when tunneling is taken into account. This was verified by calculation of the Lyapunov exponent numerically: Over any interval where $I(E)$ changes, $\lambda(E)$ was found to be zero for the expected numerical accuracy. There is no "breaking of analyticity"[12] and all eigenstates can be written in the form justified by perturbation theory:

$$f_\alpha(n)=e^{ikn}\Phi(nQ) .$$

The contribution of the mode α to χ^{+-} at $\mathbf{q}'=\mathbf{q}+n\mathbf{Q}$ is proportional to $|\Phi(nQ)|^2$.

Open orbits of the classical picture become, with the inclusion of quantum-mechanical interference, a relatively broad continuum which develops gaps from the periodicity in the x direction. The envelope Φ for states corresponding to open orbits is relatively featureless.

Another feature suggested by Fig. 1 is the special nature of the zero-energy contour, an axis of symmetry. In fact, we can explicitly determine the two degenerate zero-energy eigenstates

$$f^{\pm}_{\alpha=0}(2n)=(-1)^n\frac{C}{(W_{2n})^{1/2}} ,$$

$$f^{\pm}_{\alpha=0}(2n+1)=\pm(-1)^n\frac{C}{(W_{2n+1})^{1/2}} ,$$

where the normalization C is given by

$$C^{-2}=\frac{N}{2\pi}\left[\frac{2}{SK(Q)}\right]\int_0^{2\pi}\frac{dx}{1+\alpha\cos x+\beta\cos(2x)} .$$

Therefore there is response for vanishing energy,

$$\lim_{\omega\to 0}\mathrm{Im}\left[\frac{\chi^{+-}(\mathbf{q}',\omega)}{\omega}\right]\propto\frac{1}{[(W_n)^{1/2}]^2}\frac{1}{W_n}$$
$$=\frac{1}{[K(\mathbf{Q})-J(\mathbf{q}')]^2} .$$

Thus there is a "quasielastic" band which has maxima at Q and $2\pi-Q$ and which is relatively broad in energy, since it appears in the region of open orbits. These excitations have the greatest amplitudes where the spin density is least.

It is tempting but incorrect to identify these low-energy excitations with the phasons whose existence follows from the broken phase symmetry of the ground state [Eq. (2)]. The present operators generate *rotations* of the spin-density wave out of the easy axis and not a shift in its phase. The phason operator, in contrast, would generate low-frequency response in the *longitudinal* response function. To the order of the present calculation, fluctuations in the transverse correlation functions are not coupled to longitudinal response, although, of course, such coupling should appear at higher order. The low frequencies here appear purely because of the existence of low spin densities, and therefore restore forces in a linear approximation, close to the nodes of the spin density (2).

Strictly speaking, it is probable that all the "bands" described here are not mathematically continuous bands: It has been conjectured[10] that the spectrum of quasiperiodic operators such as (14) is a Cantor set with an infinite number of gaps, each of finite but increasingly tiny measure. There is likely, then, to be a certain vagueness in our definition of the number of bands observed, in that if we could define energy to increasingly higher accuracy we might distinguish an increasing number of distinct bands. In practice, this is not a serious constraint as the ignored structure is so fine: Operationally we have effectively included a "resolution" of the order of 10^{-3} times the couplings. This should certainly outdo any experiments.

We emphasize that numerical procedures may not distinguish the multitude of tiny gaps, but they do not miss the exponentially narrowed bands. Although the function $\lambda(E)$ may vanish on a tiny interval, over the same interval $I(E)$ changes sharply, and thus the interval over which it changes can be defined with great ease. While we expect that the *complement* of the spectrum is dense, and therefore increasingly accurate computation should reveal further gaps within a previously defined band, the same is not true of the *spectrum*: a gap should remain a gap.

RESULTS OF NUMERICAL ANALYSIS

It is only in the frequency dependence that sharp structure is displayed: the $\mathbf{q}$ dependence is smooth. This is a consequence of the fact that the states are extended in $\mathbf{q}$ space: if they were localized (positive Lyapunov exponent) the structure would be of sharply defined excitations in q with a dense set of satellites, corresponding to the point spectrum. In practice, we calculate the spectral weight of eigenmodes by the procedure outlined above with of order 10000 modes; we then determine the momentum dependence of $\chi^{+-}(\mathbf{q},\omega)$ in each frequency range by a straightforward diagonalization of a tridiagonal matrix corresponding to a few hundred modes and fold the results together. This procedure gives an accurate calculation of $\chi^{+-}(\mathbf{q},\omega)$ with modest computing time.

In Fig. 2 we show the function $\mathrm{Im}[\chi^{+-}(\mathbf{q},\omega)/\omega]$ calculated for the case of modulation with small $\mathbf{Q}$. It is verified that in addition to the smearing out of spin-wave excitations of the ferromagnetic state $\mathbf{Q}=0$, as predicted by perturbative theories,[4] there are two qualitatively new features for $\mathbf{Q}$ close to the commensurable vector $\mathbf{Q}_c$ (here 0).

(i) In the high-frequency region at least one band of response is extremely sharply defined in energy. In the limit $\delta\mathbf{Q}=|\mathbf{Q}-\mathbf{Q}_c|\to 0$ the total weight vanishes as $\delta\mathbf{Q}$, the energy width as $\exp(-\mathrm{const}/\delta\mathbf{Q})$. The frequency is comparable to that of band-edge magnons in the com-

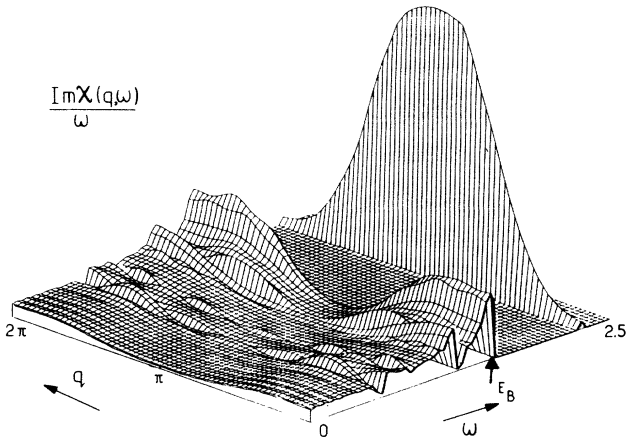

FIG. 2. Imaginary part of the dynamical susceptibility as calculated for the same parameters as Fig. 1. The energy units are such that $SK(Q)/2 \equiv 1$. The arrow labeled E_B corresponds to the energy $E_B = 1.481$ of the classical separatrix B of Fig. 1. The sharp resonance at higher frequencies is bounded by energies [2.2411, 2.2547]. The dips in intensity at lower frequency would descend to zero if the plotting resolutions were finer.

mensurable case. The momentum dependence of the dynamic susceptibility at these energies is quite characteristic: it is effectively the density of a particle in the bound states of a potential giving the closed classical orbits of Fig. 1. For the case drawn, the extreme band has a maximum at $\mathbf{q}' = \pi$; for an almost antiferromagnetically ordered case $Q \simeq \pi$, the two local maxima in $f(q)$ give a two-peaked structure for the highest frequency. We can approximate the potential close to its minima or maxima by a harmonic form. Thus as $\delta\mathbf{Q} \to 0$ the extremal bands are evenly spaced in energy, with momentum dependence corresponding to the spatial dependence of linear harmonic-oscillator wave functions. As the energy ω decreases, the bands broaden in energy and have more maxima as a function of q, corresponding to the structure of less highly bound states.

(ii) A quasielastic, i.e., low-energy, feature that has a broad maximum at the incommensurable vectors $\pm\mathbf{Q}$. The total weight vanishes as $\delta\mathbf{Q}$ and the energy width is not exponentially small. As we have stressed, these are associated with the regions of low spin density in which the linearization of the equations of motion is least likely to be accurate.

GENERALIZATIONS AND CONCLUSIONS

The model (1) studied here is rather simplified: for satisfactory application to rare-earth systems it is necessary to include terms higher than quadratic in the crystal field. The main complication is that two mechanisms induce third-, fifth-, ..., order couplings to the tight-binding model (7): (a) terms in the Hamiltonian (1) of higher order than quadratic in the spin operators, and (b) higher harmonics in the ground state (2). In addition, crystal-field terms that break axial symmetry add operators S_q^- in the equation of motion for S_q^+ and vice versa. To include cubic or hexagonal anisotropy, it is therefore necessary to formulate the tight-binding model in terms of pseudo-spin-$\frac{1}{2}$ operators. This renders numerical calculation slightly harder but no different in principle. Of course, it is quite possible that with these modifications there may emerge a pure point spectrum: This will be signaled by a positive Lyapunov exponent in the region of nonzero spectral measure.

The methods developed here are also applicable to excitations in spiral phases when "bunching" occurs, i.e., when a nonuniform turn angle is induced by applied-magnetic-field or crystal-field anisotropies. If bunching is seen in static studies, as it has been in holmium,[3] the spin-wave equations should be modified from those of the regular helix. When this is done[2,13] the transformation to a rotating frame of coordinates that successfully decouples the equations for the undistorted helix fails to separate modes of different momenta. Again the coupled set of operator equations may be brought into a form similar to that presented here and solved by the same methods. In this case preliminary calculations[13(b)] indicate that the spectrum *is* pure point.

We emphasize that the one dimensionality of the equations for the three-dimensional phases considered is a consequence of the "single-Q" ordering that couples excitations only in a one-dimensional line in the Brillouin zone of the paramagnetic phase. Higher harmonics of the ordering (2) that appear as the modulation "squares up" will not affect this effective one dimensionality, nor, indeed, the qualitative features of our results. From slight extension of the arguments given, we would expect the effect to be sharpening of the momentum dependence of the high-frequency response and diminishing of the weight of the lower-frequency response. Another generalization relevant to certain phases of neodymium is of a **Q** vector *not* parallel to a symmetry direction: this preserves one dimensionality, but introduces further incommensurable periods to the effective Hamiltonian.

The same simplification of one dimensionality will not, of course, occur for a "multiple-Q" magnetic structure for which the spin density has components of nonparallel but equivalent wave vectors **Q**. A triple-**Q** structure has been proposed, for example, for neodymium, but static diffraction experiments have, so far, failed to distinguish the possibilities because of the similarity of the diffraction patterns of a single-domain multiple-**Q** structure and a multiple-domain single-**Q** structure. The dynamics associated with the different possible orderings should be distinct. While comparison of the two possibilities by dynamic measurements would require a theory like the present one for the triple-**Q** structure, a theory that is currently lacking, it is already suggestive that the high-frequency feature (i) described above is enhanced by the effective one dimensionality. The feature should be distinctive through the fact that it occurs at a frequency that varies transverse to, but not parallel to, **Q**. This suggests that the two possibilities, single or multiple **Q**, could be separated by a dynamic measurement. Naturally, proper interpretation of experiments requires a fuller treatment of crystal-field splittings, both in the manner outlined above and by introduction of standard basis operators[14] that preserve the "excitonic" nature of magnetic excitations in the presence of large crystal-field splittings.

It is well known that modulated magnetic structures

also occur in transition metals, such as chromium. In this case the itinerancy of the electrons means that a localized-moment model as employed here is inapplicable, strictly speaking. The present calculations do show that, however, even neglecting itinerancy, the excitation spectrum of a modulated magnet is richer than hitherto assumed. While we would refrain from applying the present theory to chromium, it is clear that the recent excitations discovered[15] should be interpreted with this in mind.

In this paper we have discussed linear spin-wave equations for sinusoidally modulated structures. A number of interesting new features occur in the spectrum in addition to the broadened spin-wave bands. We emphasize that these conclusions, while exact consequences of the linearized equations of motion (5), are based on those equations and are not directly on the Hamiltonian (1). In applying the theory it is important to remember that fluctuation corrections to (5) may blur some of the sharp features we predict. Furthermore, static structures corresponding to (2) are generally stabilized by entropy considerations and are therefore stable at finite temperatures. Thus thermal fluctuations are significant and should be taken into account by including a thermal averaging in the dynamical correlation functions. While a detailed discussion of fluctuation effects is clearly beyond the scope of this paper, it is clear that a correct treatment of the linearized theory, as we have sought to present here, is the initial step in such a direction.

ACKNOWLEDGMENTS

We would like to thank the Institut Laue-Langevin for its hospitality to one of us (P.A.L.), and Nordisk Institut for Teoretisk Atomfysik (Denmark) and Risø National Laboratory for the same to the other (T.A.L.Z.), as well as our experimental colleagues for encouragement and advice, especially W. G. Stirling, K. A. McEwen, B. Lebech, H. Stanley, R. Pynn, S. Sinha, B. Rainford, and R. Currat.

---

*Present address: Serin Physics Laboratory, Rutgers University, Piscataway NJ 08854.

[1]For recent reviews of the magnetic structures of these materials, see B. Lebech, J. Appl. Phys. **52**, 2019 (1981). A more general review is S. K. Sinha, in *Handbook of Physics and Chemistry of Rare Earths,* edited by K. A. Gschneider, Jr. and L. Eyring (North-Holland, Amsterdam, 1968), Vol. 1, p. 489. Recent dynamic measurements are reported for neodymium by K. A. McEwen and W. G. Stirling, J. Magn. Magn. Mater. **30**, 99 (1982), for praseodymium by K. A. McEwen, W. G. Stirling, and C. Vettier, Physica **120B**, 152 (1982), and for $CeAl_2$ by R. Osborn, M. Loewenhaupt, B. D. Rainford, and W. G. Stirling (unpublished). These measurements are summarized by W. G. Stirling and K. A. McEwen, in *Neutron Scattering,* edited by K. Sköld and D. L. Price (Academic, New York, in press), Chap. 20.

[2]B. R. Cooper, R. J. Elliott, S. J. Nettel, and H. Suhl, Phys. Rev. **127**, 57 (1962).

[3]G. P. Felcher, G. H. Lander, T. Arai, S. K. Sinha, and F. H. Spedding, Phys. Rev. B **13**, 3034 (1976).

[4]S. H. Liu, J. Magn. Magn. Mater. **22**, 93 (1980); P. A. Lindgård, *ibid.* **31-34**, 603 (1983).

[5]See the discussion in R. J. Elliott and R. V. Lange, Phys. Rev. **152**, 235 (1966).

[6]A recent review is by R. Currat, in *Multicritical Phenomena,* edited by R. Pynn and A. J. Skjeltorp (Plenum, New York, 1984), pp. 178.

[7]P. G. Harper, Proc. Phys. Soc. London, Sect. A **68**, 874 (1955); D. R. Hofstadter, Phys. Rev. B **14**, 2239 (1976).

[8]R. J. Elliott, Phys. Rev. **124**, 346 (1961).

[9]B. R. Cooper, in *Solid State Physics,* edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic, New York, 1968), Vol. 21, p. 393, is a useful review of theory and older experiment.

[10]S. Aubry and G. André, in *Proceedings of the Israel Physical Society,* edited by C. G. Kuper (Hilger, Bristol, 1979), Vol. 3, p. 133; S. Ostlund and R. Pandit, Phys. Rev. B **29**, 1394 (1984); M. Wilkinson, Proc. R. Soc. London, Ser. A **391**, 305 (1984); recent mathematical work is reviewed by B. Simon, Adv. Appl. Math. **3**, 463 (1982).

[11]See K. Ishii, Prog. Theor. Phys. Suppl. **53**, 77 (1973), and V. I. Ossedelec, Trans. Mosc. Math. Soc. **19**, 197 (1968), for a discussion of the calculation of the Lyapunov exponent. The method implemented here is identical to that used by T. A. L. Ziman, Phys. Rev. Lett. **49**, 337 (1982), save that in that application the $\{t_n\}$ were random variables. The method of calculating the integral density of states is simply that of the Sturm sequence.

[12]See S. Aubry and G. André, in *Proceedings of the Israel Physical Society,* Ref. 10. There seems to be no reason, in principle, why localized and extended bands should not coexist at different energies; this does not occur for Harper's equation, however, because of self-duality, but apparently nor does it occur here, at least to the numerical accuracy obtained and the parameters studied. Note that throughout we identify "localized" or "extended" with the property of having a positive or a zero Lyapunov exponent, respectively. While this identification is adequate for the present purposes, the equivalence is not rigorous: See, for example, the counterexample, for special irrational periods, of S. Prange, D. Grempel, and S. Fishman, Phys. Rev. B **28**, 7370 (1983). Furthermore, for certain values of parameters and incommensurate periods, the spectra may be "exotic" (see B. Simon in Ref. 3). We do not consider such mathematical subtleties in the present treatment.

[13](a) T. Arai and G. P. Felcher, J. Phys. C **8**, 2095 (1975); (b) T. Ziman and J. Keating (unpublished).

[14]S. B. Haley and P. Erdös, Phys. Rev. B **5**, 1106 (1972); W. J. L. Buyers, T. M. Holden, and A. Perreault, *ibid.* **11**, 266 (1975).

[15]S. K. Burke, W. G. Stirling, K. R. A. Ziebeck, and J. G. Booth, Phys. Rev. Lett. **51**, 494 (1983).