Theorem on Spin Waves in Helical Spin Structures Adapted from the Goldstone Theorem

R. J. ELLIOTT*
Department of Theoretical Physics, Oxford University, Oxford, England

AND

R. V. LANGE†‡
Atomic Energy Research Establishment, Harwell, England
and
Department of Theoretical Physics, Oxford University, Oxford, England

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It is shown that rare-earth metals and other substances which display helical spin ordering have no energy gap in their spin-wave spectra even in the presence of an external magnetic field. The field is not treated in perturbation theory. Rather, the ideas of the relativistic Goldstone theorem are adapted to this problem and the result is proved by using only the linearization usual in spin-wave theories, the symmetry of the ground state, and the finite range of the exchange interaction.

1. INTRODUCTION

HELICAL, conical, and even more complex magnetic orderings first predicted by Yosimori[1] are known to exist in the rare-earth metals[2] and in some other materials like $MnAu_2$.[3] The spin-wave spectra of such structures are complicated, but their study has been begun by magnetic-resonance,[4] neutron-diffraction,[5] specific-heat,[6] and transport measurements.[7] The most detailed theoretical discussion at present published is that of Cooper *et al.*[8] They considered in detail a helical structure in the presence of a magnetic field in the plane of spin orientation. In this situation it has been shown by Nagamiya *et al.*,[9] that modifications of the helical structure occur. As the applied field is increased, small distortions of the helix are produced, giving a moment along the field. At a certain critical field H_c there is a first-order transition, accompanied in real crystals by a lattice distortion,[10] to a fan-like structure with a large moment along **H**. As the field is increased further, the angle of the fan decreases until complete ferromagnetic alignment is achieved in a second-order transition at $H=H_f$, which is approximately twice H_c. This analysis showed that the spin-wave spectrum was continuous down to zero frequency in the case of a pure helix ($H=0$) and at $H=H_f$, but that there was a finite gap and no zero-frequency modes at other fields. In recent detailed work, Thomas and Wolf[11] (and independently Nagamiya[12]) have shown this conclusion of Cooper and Elliott to be wrong.[11a] When numerical errors are corrected, $\omega=0$ to the order considered.

It therefore seems that there is a zero-frequency mode throughout the helical and fan range to the order of these calculations, and this suggests a general result,[11] independent of the perturbation methods used. This conjecture is proved in this paper by a method related to the Goldstone theorem in relativistic theory.[13]

An intuitive picture of the contents of the Goldstone theorem in a nonrelativistic context is most easily formed by considering a simple ferromagnet. The ground state of a ferromagnet is degenerate; each ground state is specified by its magnetization direction. These other ground states can be considered zero-energy excitations from any given ground state. The Goldstone-theorem technique formalizes the intuitive notion that there might be low-lying states which are arbitrarily close in energy and spin configurations to a simple rotation of the entire ferromagnet. These would be long wavelength spin waves which, over distances less than a wavelength, look like rotations of the magnetization but for which the angle of rotation varies slowly as one moves through the crystal. A direct translation of the Goldstone theorem into nonrelativistic language attempts to prove the existence of such an excitation branch with no energy gap without reference

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† National Science Foundation Post Doctoral Fellow.
‡ Present address: Brandeis University, Waltham, Massachusetts. Work supported in part under National Science Foundation Contract No. GP5374.
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to the details of the interactions. Only the degeneracy of the ground state and the existence of an appropriate conservation law, which in the ferromagnet is the microscopic spin-conservation law, are invoked. There has recently been considerable discussion[14,15] of the content of the Goldstone theorem in nonrelativistic theories, and Lange[15] has shown that the existence of an excitation mode with no energy gap can not be proved just from the symmetry conditions and conservation laws alone. The theorem can be expected to fail unless the forces are of finite range. Rigorous results are obtained in this paper by combining the ideas behind the Goldstone theorem with the explicit use of the finite range of the spin interactions.

This type of approach has been presented in a more general context in another paper.[16]

2. FORMALISM

Following the notation of Cooper and Elliott,[8] we use a model Hamiltonian

$$\mathcal{H}=-\sum_j\sum_{i\neq j}J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j-K_2\sum_i S_{i\zeta}^2+\lambda\beta H\sum_i S_{i\xi},\quad(1)$$

which has the essential features, *viz.*, exchange, anisotropy, and a field applied in the plane. ζ is chosen as the symmetry axis, ξ, η as directions in the plane perpendicular to ζ. ξ is along and η is perpendicular to $\mathbf{H}$. In the rare earths the spin arrangement is such that the moments of all the atoms in a given layer perpendicular to the symmetry axis are the same, but the moments vary in direction, and possibly slightly in size, from layer to layer. Taking the angle between the moment in the nth layer and the ξ axis as θ_n, we define a new set of axes for each layer with $\mathbf{Z}_n$ along the moment direction.

$$\begin{aligned}\mathbf{X}_n&=-\boldsymbol{\zeta},\\ \mathbf{Y}_n&=-\boldsymbol{\xi}\sin\theta_n+\boldsymbol{\eta}\cos\theta_n,\\ \mathbf{Z}_n&=\boldsymbol{\xi}\cos\theta_n+\boldsymbol{\eta}\sin\theta_n.\end{aligned}\quad(2)$$

The moment μ_n is proportional to the ground-state expectation value of $S_n{}^Z$,

$$\mu_n=-\lambda\beta\langle 0|S_n{}^Z|0\rangle=-\lambda\beta M_n,\quad(3)$$

where λ is the Landé factor taken with negative sign.

For the perfect helix, θ varies linearly with the position of the layer along the ζ axis, say, like $\kappa\zeta$. The wavelength $2\pi/\kappa$ is not in general commensurate with the lattice spacing c, i.e., $\kappa c/2\pi$ is an irrational number. The actual values of $\zeta=nc$ are discrete, but if θ is always referred to the range $-\pi$ to π, all values of θ will be found with equal probability. In the distorted spiral, θ is no longer linear, but has a periodic variation of wavelength $2\pi/\kappa$ superimposed on the linear function $\theta=\kappa\zeta$. In the fan phase, a periodic variation is found about $\theta=0$ (see Fig. 1).

Using a semiclassical approximation for the spins, the total energy of the system in this configuration is

$$E=-\sum_j\sum_{i\neq j}J_{ij}M_iM_j\cos(\theta_i-\theta_j)-\sum_i\lambda\beta H\cos\theta_i.\quad(4)$$

In equilibrium the free energy is a minimum. The entropy will, however, depend only on the μ_i and not on the θ_i. Thus one set of conditions for equilibrium is

$$\partial E/\partial\theta_i=2\sum_j J_{ij}M_j\sin(\theta_i-\theta_j)+\lambda\beta H\sin\theta_i=0.\quad(5)$$

The ground state described above is always degenerate. For the perfect helix ($\mathbf{H}=0$), this degeneracy is simply understood in terms of the invariance of the energy when an arbitrary phase angle is added to θ. It may also be considered as a movement of the spin order relative to the lattice. Looked at this way, it is obvious that a similar degeneracy will exist in the distorted structures ($\mathbf{H}\neq0$) corresponding to a movement of the curves in Fig. 1 parallel to the ζ axis. The θ_i will readjust to remain in equilibrium. If we regard θ as a continuous function of ζ, then the equilibrium condition (5) will be independent of this translation, and for later consideration we write this

$$\frac{\partial}{\partial\zeta}\left(\frac{\partial E}{\partial\theta_i}\right)=0.\quad(6)$$

When the pattern repeats exactly after, say, ν layers there is only a finite set of θ values, $\theta_1,\cdots,\theta_\nu$, which are explicitly determined by (5). The continuous function $\theta(\zeta)$ cannot then be defined, and it is not immediately clear whether there is a degeneracy in the ground state such as that given by Eq. (6). We may, however, extend the argument to cover this case. Nagamiya

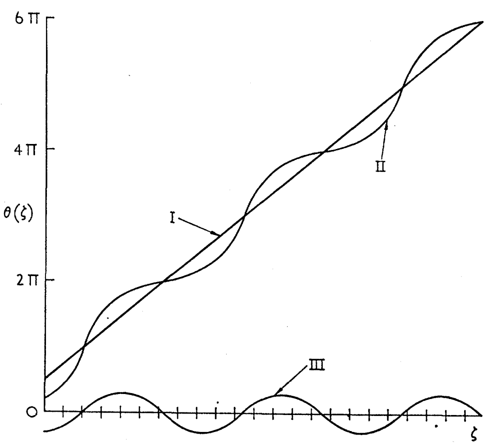

FIG. 1. The functions $\theta(\zeta)$ appropriate to various configurations are: I, helix ($H=0$; $\theta=\kappa\zeta$); II, distorted helix ($0<H<H_c$); and III, fan ($H_c<H<H_f$). The hatches on the ζ axis mark the points $\zeta=nc$. $\theta(nc)$ and $\theta'(nc)$ are the values appropriate to the nth layer. The transformations among ground states correspond to horizontal translations of these curves.

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et al.[9] showed that the repeating distance, whose inverse is $\kappa/2\pi$, varies with H. As H increases, κ changes from the pure helix value κ_0, has a discontinuous change at H_c, and then goes smoothly back to κ_0 at H_f. We can say that the slope of the mean line in region II of Fig. 1 should vary slightly with H, as will the period of the oscillations in II and III.

Thus, if we are ever at a commensurate situation we can move to an incommensurate one by a slight variation of H. In that case, $\theta(\zeta)$ is defined and we can move to a degenerate state by a translation. Reversing the variation of H will then return us to a state of the commensurate situation degenerate with the first, and different from it. We therefore conclude that the degeneracy theorem will hold for all commensurate cases which can be reached from incommensurate ones by a small change in H. This is not true when $\kappa=0$ or π/c, i.e., at the zone center or the zone boundary, corresponding to simple ferro- and antiferromagnetism.

In each of these two cases there is a single ground state when $H\neq 0$. For the ferromagnet, all the moments are along the field, for the antiferromagnet they set nearly perpendicular to the field. But in the general case of a pattern repeating after ν layers there are a degenerate set of states, obtained as outlined in the previous paragraph, or, for example, by rotating the helix relative to the field direction before it is applied. Since the translation of the helix gives a continuous degeneracy, the degeneracy is continuous in the commensurate situation. The existence of a zero-energy spin-wave mode has been confirmed by direct calculation for $\nu=3$ and 4, and the $\nu=3$ case is given in the Appendix.

3. ZERO-FREQUENCY EXCITATION

The energies of the spin-wave modes may be obtained from the equations of motion of the X and Y components of the spins. These equations can be linearized since they will always be used in operation on the ground state and the spin deviations from equilibrium may be regarded as small. For our purposes we need

$$\begin{aligned} i\hbar(\partial S_i^X/\partial t) &= [\mathcal{H},S_i^X] \\ &= \sum_j J_{ij}M_iM_j \sin(\theta_i-\theta_j)+\lambda\beta HM_i \sin\theta_i \\ &\quad -2\sum_j J_{ij}(M_jS_i^Y-M_iS_j^Y)\cos(\theta_i-\theta_j) \\ &\quad -\lambda\beta HS_i^Y\cos\theta_i+O(S_i^YS_j^Y). \end{aligned} \tag{7}$$

The terms independent of the spin operators S_i^Y vanish because of the equilibrium condition (5).

The zero-frequency excitation should be that which transforms the system among its degenerate ground states. This corresponds to a rotation of the spin about the ζ (or X) axis by an amount which corresponds to the translation of the $\theta(\zeta)$ curve. The infinitesimal transformation which generates a spin rotation equivalent to a translation of the $\theta(\zeta)$ curve an infinitesimal distance $\delta\zeta$ is

$$U(\delta\zeta)=1+i/\hbar\sum_n \theta_n' S_n^X\delta\zeta, \tag{8}$$

where θ_n' is the derivative of the curve in Fig. 1 at the nth lattice-plane position. That this transforms the ground state into another ground state can be checked by summing over Eq. (7) and operating on the ground state.

$$\begin{aligned} i\hbar\left(\frac{\partial}{\partial t}\right)&\sum_n \theta_n' S_n^X|0\rangle \\ &=2\sum_{mn} J_{mn}\theta_n'(M_mS_n^Y-M_nS_m^Y)\cos(\theta_m-\theta_n)|0\rangle \\ &\qquad -\lambda\beta\sum_n S_n^Y\theta_n' H\cos\theta_n|0\rangle, \\ &=2\sum_n\Big[\sum_m J_{mn}M_m\cos(\theta_m-\theta_n)(\theta_n'-\theta_m') \\ &\qquad -\lambda\beta H\theta_n'\cos\theta_n\Big]S_n^Y|0\rangle, \\ &=2\sum_n\frac{\partial}{\partial\zeta}\left(\frac{\partial E}{\partial\theta_n}\right)S_n^Y|0\rangle, \\ &=0. \end{aligned} \tag{9}$$

Thus the communtator $[\mathcal{H},U(\delta\zeta)]$ gives zero when operating on the ground state and therefore the operator $U(\delta\zeta)$ transforms the ground state into another state of the same energy. In other words, the spin wave excited by $\sum_n \theta_n' S_n^X$ has zero energy.

It remains to show that this mode is not isolated but part of a continuum. To do this one might construct modes which also have a wave-like variation $e^{i\mathbf{k}\cdot\mathbf{R}_n}$ across the crystal, and consider the limit $\mathbf{k}\to 0$. This is equivalent to solving the general problem as attempted in Refs. 8 and 11, and is too difficult in the general case. Instead, we shall use a method related to the Goldstone theorem.

4. PROOF OF NO ENERGY GAP

S_n^X and S_n^Y are conjugate operators so that in the ground state, for all times t,

$$\langle 0|[S_n^X(t),S_m^Y(t)]|0\rangle = i\langle 0|S_n^Z(t)|0\rangle\delta_{nm}. \tag{10}$$

In the present situation, $\theta_n' S_n^X$ is the operator of interest, and it has this same commutation relation with S_m^Y/θ_m'. In the fan case, however, θ_m' can become zero, so that this operator is not a satisfactory conjugate to $\theta_m' S_m^X$. Instead, we use the pair

$$\begin{aligned} \psi_n(t) &= \theta_n' S_n^X(t), \\ \phi_m(t) &= \sum_{l\sim m}\theta_l' S_l^Y(t)\Big/\sum_{l\sim m}(\theta_l')^2, \end{aligned} \tag{11}$$

where $\sum_{l\sim m}$ is a summation of l over a finite region about m. Then

$$\sum_n e^{-i\mathbf{k}\cdot(\mathbf{R}_n-\mathbf{R}_m)}\langle 0|[\psi_n(t),\phi_m(t)]|0\rangle = i\sum_{l\sim m}(\theta_l')^2 M_l e^{-i\mathbf{k}\cdot(\mathbf{R}_l-\mathbf{R}_m)}/\sum_{l\sim m}(\theta_l')^2. \quad (12)$$

The region in the summation is chosen to be finite, but large enough so that

$$\sum_{l\sim m}(\theta_l')^2 M_l/\sum_{l\sim m}(\theta_l')^2 = M, \quad (13)$$

where M is an average moment essentially independent of m. If r is the linear dimension of the region, then for $kr\ll 1$

$$\sum_n e^{-i\mathbf{k}\cdot(\mathbf{R}_n-\mathbf{R}_m)}\langle 0|[\psi_n(t),\phi_m(t)]|0\rangle = iM. \quad (14)$$

The function

$$L(\mathbf{k},\omega) = \sum_n e^{-i\mathbf{k}\cdot(\mathbf{R}_n-\mathbf{R}_m)} L_{nm}(\omega), \quad (15)$$

where

$$L_{nm}(\omega) = \int dt e^{i\omega t}\langle 0|[\psi_n(t),\phi_m(0)]|0\rangle, \quad (16)$$

will give the power spectrum in frequency of excitations of wave vector $\mathbf{k}$. We know from the previous discussion of the zero-frequency mode that when $\mathbf{k}=0$, $L(0,\omega)$ is proportional to $\delta(\omega)$. The constant of proportionality M can be determined from Eq. (14), since for any $\mathbf{k}$ such that $kr\ll 1$,

$$\int \frac{d\omega}{2\pi} L(k,\omega) = iM. \quad (17)$$

We need to show that this $\delta(\omega)$ form can be obtained in the limit $\mathbf{k}\to 0$. Instead of considering finite $\mathbf{k}$ explicitly, we examine $\sum_{n\in V} L_{nm}(\omega)$, a summation over sites in a volume V, centered at site m. If there are N sites in the entire crystal, then

$$\sum_{n\in V} L_{nm}(\omega) = \frac{N_V}{N} L(0,\omega) + \frac{1}{N}\sum_k{}' \left[L(\mathbf{k}\omega)\times\sum_{n\in V} e^{i\mathbf{k}\cdot(\mathbf{R}_n-\mathbf{R}_m)}\right], \quad (18)$$

where N_V is the number of sites in volume V and $\sum'$ has $\mathbf{k}=0$ excluded. If we first let $N\to\infty$ while V and therefore N_V remain finite, the first term in Eq. (18) vanishes and

$$\lim_{N\to\infty}\sum_{n\in V} L_{nm}(\omega) = \int \frac{d^3k}{(2\pi)^3}\bar{L}(\mathbf{k}\omega)\Delta_V(k). \quad (19)$$

The $\mathbf{k}$ summation has become an integral over the product of $\bar{L}(\mathbf{k}\omega)$, defined by

$$\bar{L}(k\omega) = L(\mathbf{k}\omega), \qquad k\neq 0$$
$$= \lim_{\mathbf{k}\to 0} L(\mathbf{k}\omega), \quad \mathbf{k}=0, \quad (20)$$

and $\Delta_V(\mathbf{k})$ defined by

$$\Delta_V(\mathbf{k}) = \lim_{N\to\infty}\sum_{n\in V} e^{i\mathbf{k}\cdot(\mathbf{R}_n-\mathbf{R}_m)}\Omega, \quad (21)$$

where Ω is the volume of a cell of the lattice. By using the function $\bar{L}$ which is regular at zero $\mathbf{k}$, we assure that the integral in Eq. (19) is the proper limiting form of $\sum'$ without invoking any assumptions about the relationship between $L(\mathbf{k}\omega)$ for $\mathbf{k}$ finite and $L(0,\omega)$. Finally, we let the volume V tend to infinity. Since

$$\lim_{V\to\infty}\Delta_V(\mathbf{k}) = (2\pi)^3\delta(\mathbf{k}), \quad (22)$$

we get

$$\lim_{V\to\infty}\lim_{N\to\infty}\sum_{n\in V} L_{nm}(\omega) = \lim_{k\to 0} L(k\omega). \quad (23)$$

It may be noted that if the limits are taken the other way round the result is

$$\lim_{N\to\infty}\lim_{V\to N\Omega}\sum_{n\in V} L_{nm}(\omega) = L(0,\omega). \quad (24)$$

The summation will be analyzed through its moments which are given by

$$\int \frac{d\omega}{2\pi}\omega^P \sum_{n\in V} L_{n0}(\omega) = \langle 0|\sum_{n\in V}\left[(i)^P\frac{d^P\psi_n(0)}{dt^P}, \phi_0(0)\right]|0\rangle. \quad (25)$$

The summation is over a volume V larger than (and containing) the region introduced in the definition of ϕ_0, so that the special case for $P=0$ is

$$\int \frac{d\omega}{2\pi}\sum_{n\in V} L_{n0}(\omega) = iM. \quad (26)$$

The first derivative is given by

$$i\hbar\frac{\partial}{\partial t}\sum_{n\in V}\psi_n(t) = 2\sum_{n\in V, m\notin V} J_{nm}\cos(\theta_n-\theta_m) \times (M_m\theta_m' S_n^Y - M_n\theta_n' S_m^Y). \quad (27)$$

The right-hand side of Eq. (27) consists of surface terms, in that if the exchange interaction J_{nm} has finite range ρ, then all the spin operators refer to sites within ρ of the surface of V. In a corresponding expression for the nth derivative of $\sum_{n\in V}\psi_n(t)$, the right-hand side would refer to sites within $n\rho$ of the surface. Therefore, if the surface is everywhere at least a distance R from the sites of the spin operators in ϕ_0 we find that

$$\int \frac{d\omega}{2\pi}\omega^P \sum_{n\in V} L_{n0}(\omega)=0 \quad \text{for} \quad 0<\rho<R/P, \tag{28}$$

since the equal-time commutator of spin operators on different sites is zero. In the $V\to\infty$ limit we get

$$\int \frac{d\omega}{2\pi}\omega^P \lim_{V\to\infty}\lim_{N\to\infty}\sum_{n\in V} L_{n0}(\omega)=0 \tag{29}$$

for all P except $P=0$, and therefore

$$\lim_{\mathbf{k}\to 0} L(\mathbf{k}\omega)=iM\delta(\omega), \tag{30}$$

where Eq. (26) has been used to set the multiplicative constant.

Thus the zero-frequency mode is not an isolated one but part of a continuous spectrum. The power spectrum at zero **k** is the zero-**k** limit of the power spectrum at finite **k**. The spin-wave spectrum reaches down to zero frequency with no gap irrespective of the external field **H**. By the discussion at the end of Sec. 2, the result may be extended to commensurate spin arrangements other than ferro- and simple antiferromagnetism. As κ varies with H and T in the rare earths, it passes continuously through such arrangements. It is satisfying that the theorem holds without discontinuities at these points.

We might add that similar results will hold for a conical ordering, such as is found in Er, or may be induced in a helical ordering if a component of the magnetic field is along ζ. They will also hold in the presence of anisotropy in the plane. If this is n-fold, it will impose a further variation on $\theta(\zeta)$ periodic in $2\pi/n$, but the argument will go through as before.

When any of these types of magnetic phase are found at low temperatures, the specific heat and resistivity from spin-wave mechanisms will have a simple power-law variation with T, and this fact will not be affected by the application of a magnetic field.

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APPENDIX

For a pattern repeating after ν layers, we need only consider those motions where $S_i^X=S_{i+\nu}^X$. There are then ν equations of type (7) as well as ν equations of type (5). They will have the same form with

$$\sum_m J_{i,j+\nu n}=J_{ij}. \tag{A1}$$

Writing the S^X as a column vector, Eq. (7) may be summarized as a matrix relation

$$\hbar\omega S^X=AS^Y. \tag{A2}$$

From the equation of motion for S_i^Y, a similar relation

$$\hbar\omega S^Y=BS^X \tag{A3}$$

may be obtained. The condition for a zero root is $\det(AB)=0$, and hence either $\det A=0$ or $\det B=0$. Since B will involve the anisotropy energy, $\det A=0$ is the only possibility.

For $\nu=3$ the fact that $\det A=0$ may be demonstrated by manipulation. From Eq. (A1),

$$J_{12}=J_{23}=J_{31}=J. \tag{A4}$$

A typical diagonal element of A is

$$\begin{aligned} J[\cos(\theta_1-\theta_2)+\cos(\theta_1-\theta_3)]+\lambda\beta H\cos\theta_1 \\ =\cos\theta_1(J\cos\theta_2+J\cos\theta_3+H) \\ +J\sin\theta_1(\sin\theta_2+\sin\theta_3). \end{aligned} \tag{A5}$$

But from the condition (5),

$$J[\sin(\theta_1-\theta_2)+\sin(\theta_1-\theta_3)]+\lambda\beta H\sin\theta_1=0. \tag{A6}$$

By summing the three similar conditions of this type,

$$\sum_i \sin\theta_i=0. \tag{A7}$$

Using Eqs. (A6) and (A7) gives a value $-J$ for the right-hand side of Eq. (A5). A then has the form

$$\begin{bmatrix} 1 & \cos(\theta_1-\theta_2) & \cos(\theta_1-\theta_3) \\ \cos(\theta_2-\theta_1) & 1 & \cos(\theta_2-\theta_3) \\ \cos(\theta_2-\theta_1) & \cos(\theta_3-\theta_2) & 1 \end{bmatrix}, \tag{A8}$$

and the determinant is readily seen to be zero. From the minors, the relative displacements in the zero-frequency mode are

$$S_1^X:S_2^X:S_3^X=\sin(\theta_2-\theta_3):\sin(\theta_3-\theta_1):\sin(\theta_1-\theta_2). \tag{A9}$$

Unfortunately, it has not proved possible to generalize this calculation to any value of ν. The special properties of this case which make this calculation simple to not appear to be closely related to the general arguments used in this paper.