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Phenomenological theory of longitudinal spin fluctuations in $CsNiCl_3$

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The recent observation of longitudinal spin excitations at low temperatures in the spin-1 quasi-one-dimensional antiferromagnet $CsNiCl_3$ is contrary to expectations based on conventional spin-wave theory. Affleck [Phys. Rev. Lett. **62**, 474 (1989)] has proposed a field-theory model which attributes this unusual gap-mode fluctuation to quantum effects resulting from the Haldane conjecture. We demonstrate here that the field-theory results are reproduced by a phenomenological Lagrangian formulation of spin waves, in the spirit of Landau and Lifshitz, that has existed in the literature for many years.

Experimental investigation into the spin dynamics of the quasi-one-dimensional, spin-1 antiferromagnet $CsNiCl_3$ has been stimulated by its candidacy for the observation of effects associated with the Haldane conjecture.[1-4] Within the framework of conventional spin-wave theory of local-moment systems, only fluctuations transverse to long-range magnetic order occur. It is thus surprising that polarized neutron-diffraction measurements in the low-temperature ordered phase of $CsNiCl_3$ give clear indication of longitudinal (amplitude modulated) spin excitations. Affleck[5] has recently shown, however, that this unusual mode follows from a $(1+1)$-dimensional field-theory-based model which provides strong support for the existence of a Haldane gap in this hexagonal insulator. It is the purpose of this work to demonstrate that a longitudinal mode $\omega_L(\mathbf{q})$ of precisely the same form as derived from field theory (in addition to conventional transverse modes) follows from a classical formalism describing spin excitations that has existed in the literature for twenty years.[6-9] Both models are based on a Lagrangian approach, with $\mathcal{L}[\phi]$ being used in Ref. 5 [where $\phi(\mathbf{r})$ is the field variable] and $\mathcal{L}[\mathbf{s}]$ forms the basis of the present theory [where $\mathbf{s}(\mathbf{r})$ is the spin density]. Differences arise in the physical interpretation of some of the parameters which appear in each model.

The magnetic Ni^{2+} ions form a simple hexagonal lattice with strong antiferromagnetic coupling along the c-axis chains. Although weak, interchain (antiferromagnetic) interactions are sufficiently strong to stabilize three-dimensional long-range magnetic order at $T_{N1}=4.85$ K, characterized by a period-2 modulation along the c axis and a period-3 structure in the basal plane, with a wave vector $\mathbf{Q}$ given by

$$\mathbf{Q}=\pm(4\pi/3a)\hat{\mathbf{x}}+(\pi/c)\hat{\mathbf{z}}\,. \tag{1}$$

A result of weak axial anisotropy is the occurrence of a second transition[10] at $T_{N2}=4.40$ K. The Hamiltonian commonly used to describe this system can be written as[11]

$$\mathcal{H}=J_{\parallel}\sum_{\langle i,j\rangle}\mathbf{S}_i\cdot\mathbf{S}_j+J_{\perp}\sum_{\langle k,l\rangle}\mathbf{S}_k\cdot\mathbf{S}_l+D\sum_i(S_i^z)^2\,, \tag{2}$$

where $J_{\parallel}>0$ and $J_{\perp}>0$ represent nearest-neighbor exchange interactions along the c axis and in the basal plane, respectively, $D<0$ is the single-ion anisotropy, and $J_{\parallel}\gg J_{\perp},|D|$. [As noted in Ref. 11, our definitions of intrachain and interchain coupling are related to those of Refs. 1-5 (J,J') by $J_{\parallel}=4J$ and $J_{\perp}=4J'$.] At very low temperatures (where the effects of D are small), the magnetic order is close to that of a 120° spin structure with moments lying in a plane containing the c axis. It is useful to note that such a structure can be characterized as a helically polarized spin density.[12,13] We will adopt here the simplification used by Affleck[5] of setting $D=0$ (also see Ref. 1).

Conventional spin dynamics are governed by the torque equation[14] (omitting relaxation effects)

$$d\mathbf{s}(\mathbf{r})/dt=\gamma[\mathbf{s}(\mathbf{r})x\mathbf{h}^e(\mathbf{r})]\,, \tag{3}$$

where γ is the gyromagnetic ratio, $\mathbf{h}^e(\mathbf{r})$ is the effective field,

$$\mathbf{h}^e(\mathbf{r})=-\delta F/\delta\mathbf{s}(\mathbf{r})\,, \tag{4}$$

and $F[\mathbf{s}(\mathbf{r})]$ is the free energy. This formalism follows the spirit of the Landau description of phase transitions. These relations can also be derived from the quantum-mechanical equation of motion for the magnetization operator[15] (based on a Hamiltonian formalism) in the semiclassical approximation (replacing F by the energy E). Only transverse spin waves can result from this approach. A more phenomenological description of magnetic excitations which has features in common with classical mechanics is based on a Lagrangian formalism,[6-9] $\mathcal{L}=T-V$ where $T[\mathbf{s}(\mathbf{r})]$ represents the kinetic energy and, as an extension from basic concepts of classical and statistical mechanics, the potential energy $V[s(\mathbf{r})]$ is taken to be of the form given by the Landau-type free energy. Both V and F have the same structure, as noted in Ref. 5. The equations of motion are of the standard form

$$\delta\mathcal{L}/\delta s_a=\frac{d}{dt}\delta\mathcal{L}/\delta\dot{s}_a\,, \tag{5}$$

with $\dot{s}_a=ds_a/dt$. The present study was motivated by the work of Izyumov and Laptev[9] who use this approach to investigate the spin dynamics of incommensurate helical magnetic structures and find a longitudinal-mode excitation (as well as conventional transverse modes). We

adopt here their expression for $T[\mathbf{s}(\mathbf{r})]$ and a Landau free energy derived from a mean-field treatment of the Hamiltonian (2).

Consider first the Landau free energy and equilibrium magnetic structure. An expression for F derived from a general Heisenberg Hamiltonian

$$\mathcal{H}=\tfrac{1}{2}\sum_{i,j}J(\mathbf{R}_i-\mathbf{R}_j)\mathbf{S}_i\cdot\mathbf{S}_j\,, \tag{6}$$

within the molecular-field approximation follows from the method of Ref. 16 (also see Ref. 17). The result, to fourth order in the spin density, can be expressed as

$$F=1/(2V^2)\int d\mathbf{r}\,d\mathbf{r}'A(\mathbf{r}-\mathbf{r}')\mathbf{s}(\mathbf{r})\cdot\mathbf{s}(\mathbf{r}') +B/(4V)\int d\mathbf{r}[\mathbf{s}(\mathbf{r})\cdot\mathbf{s}(\mathbf{r})]^2 \tag{7}$$

with

$$A(\mathbf{r})=aT+J(\mathbf{r}),\quad B=bT\,, \tag{8}$$

where $a=\frac{3}{2}$ and $b=\frac{9}{16}$ for spin-1 systems. The long-range magnetic order of $CsNiCl_3$ can be well described by a single Fourier component of the spin density[12,13]

$$\mathbf{s}(\mathbf{r})=(V/N)\sum_{\mathbf{R}}\rho(\mathbf{r})\delta(\mathbf{r}-\mathbf{R})\,, \tag{9}$$

$$\rho(\mathbf{r})=\mathbf{S}e^{i\mathbf{Q}\cdot\mathbf{r}}+\mathbf{S}^*e^{-i\mathbf{Q}\cdot\mathbf{r}}\,. \tag{10}$$

With these expressions, the free-energy simplifies to the result

$$F=A_{\mathbf{Q}}S^2+BS^4+\tfrac{1}{2}B|\mathbf{S}\cdot\mathbf{S}|^2\,, \tag{11}$$

where $S^2=\mathbf{S}\cdot\mathbf{S}^*$ and $A_{\mathbf{Q}}=a(T-T_{\mathbf{Q}})$. With nearest-neighbor interactions only, as described by (2) (and with $D=0$), one finds[12]

$$-aT_{\mathbf{Q}}=J_{\mathbf{Q}}=2J_{\parallel}\cos(cQ_z)+2J_{\perp}f_{\mathbf{Q}}\,, \tag{12}$$

$$f_{\mathbf{Q}}=\cos(\alpha Q_x)+2\cos(\tfrac{1}{2}\alpha Q_x)\cos(\mathcal{A}Q_y)\,, \tag{13}$$

where $\mathcal{A}=(\sqrt{3}/2)\alpha$. For $J_{\parallel},J_{\perp}>0$, the free energy is minimized by $\mathbf{Q}$ given in (1), so that the Néel temperature is expressed by

$$T_N=T_Q=(2J_{\parallel}+3J_{\perp})/a\,. \tag{14}$$

The polarization of the vector $\mathbf{S}$ is determined by the last term in (11) so that $\mathbf{S}\cdot\mathbf{S}=0$ is the preferred configuration since $B>0$. This describes a helical spin density,[10,12,13] e.g.,

$$\mathbf{S}=S(\hat{\mathbf{x}}+i\hat{\mathbf{z}})/\sqrt{2}\,. \tag{15}$$

Effects arising from the axial anisotropy, not considered here, are discussed in Refs. 10 and 13.

For later purposes it is convenient at this point to derive results for the static inverse susceptibility $\bar{\bar{R}}(\mathbf{q},\mathbf{q}')=\bar{\bar{\chi}}(\mathbf{q},\mathbf{q}')^{-1}$ following the method described in Ref. 18. This is accomplished by expanding the free-energy functional of the spin density away from equilibrium $[\rho^0(\mathbf{r})]$ to second order. Formally, one writes

$$\rho(\mathbf{r})=\sum_{\mathbf{q}}\mathbf{S}_{\mathbf{q}}e^{i\mathbf{q}\cdot\mathbf{r}}\,, \tag{16}$$

with $\mathbf{S}_{\mathbf{q}}=\mathbf{S}^0_{\alpha}+\tilde{\mathbf{S}}_{\alpha}$, and

$$F=F^0+\tfrac{1}{2}\sum_{\alpha,\beta}\sum_{i,j}\tilde{S}_{\alpha\mathbf{q}}R_{\beta\alpha}(\mathbf{q}',\mathbf{q})\tilde{S}_{\beta\mathbf{q}'}\,. \tag{17}$$

The susceptibility of a helical spin structure can be diagonalized by choosing a rotating (orthonormal) coordinate system,[18,19] in the present case as

$$\epsilon_+(\mathbf{r})=[\mathbf{p}_1e^{i\mathbf{Q}\cdot\mathbf{r}}+\mathbf{p}_2e^{-i\mathbf{Q}\cdot\mathbf{r}}]/\sqrt{2}\,, \tag{18}$$

$$\epsilon_-(\mathbf{r})=i[\mathbf{p}_1e^{i\mathbf{Q}\cdot\mathbf{r}}-\mathbf{p}_2e^{-i\mathbf{Q}\cdot\mathbf{r}}]/\sqrt{2}\,, \tag{19}$$

$$\epsilon_0=\mathbf{p}_3\,, \tag{20}$$

with

$$\mathbf{p}_1=(\hat{\mathbf{x}}+i\hat{\mathbf{z}})/\sqrt{2},\quad \mathbf{p}_2=\mathbf{p}_1^*,\quad \mathbf{p}_3=\hat{\mathbf{y}}\,. \tag{21}$$

Note that the equilibrium magnetization can be written as $\rho^0(\mathbf{r})=\sqrt{2}S\epsilon_+(\mathbf{r})$. The susceptibility expressed in these coordinates ($\mu,\nu=+,-,0$) is straightforward to derive, with the result $\chi_{\mu\nu}(\mathbf{q},\mathbf{q}')=\chi_{\mu}(\mathbf{q})\delta_{\mu,\nu}\Delta_{\mathbf{q},\mathbf{q}'}$, where $\chi_{\mu}(\mathbf{q})=R_{\mu}(\mathbf{q})^{-1}$ and

$$R_+(\mathbf{q})=A_{\mathbf{Q}}+4u_{\mathbf{q}}+6BS^2\,, \tag{22}$$

$$R_-(\mathbf{q})=A_{\mathbf{Q}}+4u_{\mathbf{q}}+2BS^2\,, \tag{23}$$

$$R_0(\mathbf{q})=A_{\mathbf{Q}}+4v_{\mathbf{q}}+2BS^2\,, \tag{24}$$

with

$$4u_{\mathbf{q}}=\tfrac{1}{2}(A_{\mathbf{q}+\mathbf{Q}}+A_{\mathbf{q}-\mathbf{Q}})-A_{\mathbf{Q}} =2J_{\parallel}[1-\cos(cq_z)]+J_{\perp}(3-f_{\mathbf{q}})\,, \tag{25}$$

$$4v_{\mathbf{q}}=A_{\mathbf{q}}-A_{\mathbf{Q}}=2J_{\parallel}[1+\cos(cq_z)]+J_{\perp}(3+2f_{\mathbf{q}})\,, \tag{26}$$

and $f_{\mathbf{q}}$ given by (13). Noting that $u_q\sim q^2$ at small q and that $S^2=-A_Q/(2B)$ for $T<T_N$, it can be seen that (22)–(24) represent the usual mean-field expressions for the static susceptibility in the ordered phase:[18]

$$R_+=2a(T_N-T)+4u_{\mathbf{q}},\quad R_-=4u_{\mathbf{q}},\quad R_0=4v_{\mathbf{q}}\,. \tag{27}$$

It is instructive to consider the spin excitation behavior resulting from the Landau-Lifshitz torque equations (3) and (4). With the inverse susceptibility defined by (17), the Fourier transform of these equations take the convenient form[18]

$$d\tilde{\mathbf{S}}_{\mathbf{q}}/dt=\gamma\sum_{\mathbf{q}'}(\mathbf{S}^0_{\mathbf{q}'}x\mathbf{H}^e_{\mathbf{q}-\mathbf{q}'})\,, \tag{28}$$

$$H^e_{\mu\mathbf{q}}=-\sum_{\mu}\sum_{\mathbf{q}'}\tilde{S}_{\nu\mathbf{q}'}R_{\nu\mu}(\mathbf{q},\mathbf{q}')\,. \tag{29}$$

Using results derived in the previous sections for $CsNiCl_3$ leads to the following equations of motion

$$\dot{\tilde{S}}_{+\mathbf{q}}=0\,, \tag{30}$$

$$\dot{\tilde{S}}_{-\mathbf{q}}=\sqrt{2}\gamma SR_0(\mathbf{q})\tilde{S}_{0\mathbf{q}}\,, \tag{31}$$

$$\dot{\tilde{S}}_{0\mathbf{q}}=-\sqrt{2}\gamma SR_-(\mathbf{q})\tilde{S}_{-\mathbf{q}}\,, \tag{32}$$

and the resulting frequency dispersion

$$\omega^2=2\gamma^2S^2R_-(\mathbf{q})R_0(\mathbf{q})=32\gamma^2S^2u_{\mathbf{q}}v_{\mathbf{q}}\,. \tag{33}$$

In the zero-temperature limit where $2\sqrt{2}S=1$, this result (using $\gamma=1$) becomes $\omega^2=4u_{\mathbf{q}}v_{\mathbf{q}}$, which is identical to the conventional spin-wave dispersion calculated in Ref. 1 (also see Refs. 19 and 20).

Clearly, from (30)-(32) and the basis-vector definitions (18)-(20), there is no longitudinal-mode excitation within this formalism. The coupled transverse modes correspond to a rotation of the spin density in the xz plane ($\tilde{S}_-$) and a canting out of this plane ($\tilde{S}_0$), where $\tilde{S}_-/\tilde{S}_0 \sim (v_{\mathbf{q}}/u_{\mathbf{q}})^{1/2}$. Since $J_\perp \ll J_\parallel$, it becomes clear from the expressions (25), (26), and (33) that the rotation mode is dominant at $q_z \sim 0$ with a frequency

$$\omega_- \cong [(J_\parallel q_z c)^2 + J_\parallel J_\perp (3-f_{\mathbf{q}})]^{1/2} \tag{34}$$

and that for $q_z \sim \pi/c$, the canting mode is strongest with a frequency

$$\omega_0 \cong [(J_\parallel \bar{q}_z c)^2 + J_\parallel J_\perp (3+2f_{\mathbf{q}})]^{1/2}, \tag{35}$$

where $\bar{q}_z = q_z - \pi/c$. These are the transverse modes described by Affleck's model.[5]

Finally, we consider the spin excitations resulting from the Lagrangian formalism. The kinetic energy used by Izyumov and Laptev[9] for helical magnetic structures with spins lying in the xz plane is written

$$T = \mu \int d\mathbf{r} \dot{s}^2(\mathbf{r}) + \sigma \int d\mathbf{r} s_y(\mathbf{r}) \times [s_x(\mathbf{r})\dot{s}_z(\mathbf{r}) - \dot{s}_x(\mathbf{r}) s_z(\mathbf{r})], \tag{36}$$

where μ and σ are *a priori* unknown parameters. The first term is analogous to the kinetic energy of classical mechanics and appears in Affleck's model as $1/(2\nu)\dot{\phi}^2$, with the parameter ν *a posteriori* identified as $4J=J_\parallel$. From the analogy with classical mechanics, it can be expected that the coefficient of $\dot{s}^2$ is related to the "mass" $\sim 1/J$ of spin excitations.[21] The second term (which does not appear in Affleck's model) was inspired by the work of Dzyaloshinskii and Kukharenko[7] who introduce terms of this type (linear in d/dt) using symmetry arguments and guidance from Onsager's theory of thermodynamic fluctuations.[22] As noted in Ref. 9, and as is demonstrated below, the σ term is necessary in order to reproduce the transverse spin-wave results of the torque equations; this is achieved by taking $\sigma=(2\gamma S^2)^{-1}$. We adopt here the pedantic approach of investigating the consequences of this model kinetic energy applied to $CsNiCl_3$ without further justification.

The equations of motion (5), using $\mathcal{L}=T-F$ with T given by (36) and F given by (7), reduce to the following (linearized) form in the rotating coordinate system

$$2\mu\ddot{\tilde{S}}_{+\mathbf{q}} = -R_+(\mathbf{q})\tilde{S}_{+\mathbf{q}}, \tag{37}$$

$$2\mu\ddot{\tilde{S}}_{-\mathbf{q}} + \sqrt{2}\sigma S\dot{\tilde{S}}_{0\mathbf{q}} = -R_-(\mathbf{q})\tilde{S}_{-\mathbf{q}}, \tag{38}$$

$$2\mu\ddot{\tilde{S}}_{0\mathbf{q}} - \sqrt{2}\sigma S\dot{\tilde{S}}_{-\mathbf{q}} = -R_0(\mathbf{q})\tilde{S}_{0\mathbf{q}}, \tag{39}$$

giving a longitudinal mode ($\tilde{S}_+$) not coupled to the transverse modes, as in the field-theory model. The associated amplitude-mode frequency is given by

$$\omega_+^2 = 1/(2\mu)R_+(\mathbf{q}) = 1/(2\mu)[-2A_{\mathbf{Q}} + 4u_{\mathbf{q}}], \tag{40}$$

which has a form ($\omega^2 \sim \chi^{-1}$) also encountered in the study of structural phase transitions.[23] Identification with Affleck's model is made by putting $1/(2\mu)=\nu$ and by noting that the field-theory Lagrangian contains a mass term $(\Delta^2/2\nu)\phi_i^2$ for a *single* c-axis chain i. Comparison with the free energy (7)-(14) then leads to the relation

$$A_{\mathbf{Q}} + 3J_\perp = \Delta^2/\nu. \tag{41}$$

Using this result, the longitudinal frequency becomes

$$\omega_+^2 \cong (\nu q_z c)^2 + \nu J_\perp (9 - f_{\mathbf{q}}) - 2\Delta^2, \tag{42}$$

for $q_z \sim 0$. This is precisely the amplitude-mode frequency ω_L of Affleck's model. Within the context of field-theory, however, the quantity Δ is related to the Haldane gap of an isolated chain. Note from (40) and (25) that ω_+ has its minimum value at $q=0$ so that the gap (for the three-dimensional system) can be expressed as

$$\Delta_+^2 = 2\nu a(T_N - T), \tag{43}$$

showing the decrease in Δ_+ as the Néel temperature is approached. This has been discussed by Affleck and observed experimentally by Steiner *et al.*[3] At $T=1.5$ K, this expression (with $\nu=4J$ and $T_N=4.85$ K) gives $\Delta_+=0.538$ THz, in poor agreement with the experimental value[1] of 0.19 THz. Note, however, that (43) is based on an expansion of the Landau free energy to low order in $\mathbf{s}(\mathbf{r})$, an approximation which can be expected to break down at temperatures well below T_N. At higher temperatures, the omission of uniaxial anisotropy from the present model will be important.

The frequencies of the coupled transverse-mode excitations from (38) and (39) are determined by

$$(R_- - 2\mu\omega^2)(R_0 - 2\mu\omega^2) = 2\sigma^2 S^2 \omega^2. \tag{44}$$

From this expression, it is clear that the torque-equation result (33) is reproduced by setting $\mu=0$ and $\sigma=(2\gamma S^2)^{-1}$. With $\sigma=0$ and $\mu=1/(2\nu)$, however, the transverse modes $\omega_-^2=\nu R_-$ and $\omega_0^2=\nu R_0$ are obtained, which reproduce the field-theory results (34) and (35) (valid only for small q_z). Evidently, our model with $\sigma=0$ closely resembles, in form, the model proposed by Affleck. It is clear that the σ term must be present in the kinetic energy in order to obtain a proper description of transverse spin waves in regions of $\mathbf{q}$ space away from the restricted values of q_z treated by the field-theory model.[1]

It would, of course, be more satisfying to construct a derivation of the equations of motion (37)-(39) starting from the Hamiltonian (2). This appears to be a formidable task, an observation which served to motivate Affleck's, and the present, phenomenological approach.

In conclusion, it has been demonstrated by this work that a classical Lagrangian formulation of spin excitations for spiral spin systems, exciting in the literature for some time, reproduces the results of Affleck's field-theory based model[5] for spin-waves in $CsNiCl_3$. In particular, the nonconventional longitudinal mode resulting from each model was shown to have a frequency dispersion of identical form. This result is not surprising in retrospect as both models are based on Lagrangians of similar structure. Differences lie in the physical interpretation of some of the terms in the two model Lagrangians used. Much of

Affleck's theory was inspired by quantum effects responsible for the Haldane gap in one-dimensional spin-1 systems, whereas the present model is motivated purely on the basis of a general classical Lagrangian whose lowest-order terms are justified by symmetry arguments. Within the present classical approach, the gap (43) exists even for one-dimensional spin-$\frac{1}{2}$ systems, in contrast with the Haldane conjecture. The good quantitative agreement between experimental results and the field-theory prediction is strong evidence for the existence of the Haldane gap in $CsNiCl_3$. It is nevertheless of interest that amplitude-mode fluctuations in localized spin systems are not necessarily entirely a consequence of unusual quantum states.

Note added. Results of an unconventional quantum spin-wave theory for one-dimensional antiferromagnets have recently been reported[24] which yield an energy gap for both integer and half-odd-integer spin systems.

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