Theory of near-zero-wave-vector neutron scattering in Haldane-gap antiferromagnets

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One-dimensional integer-spin Heisenberg antiferromagnets have disordered ground states and a gap to a triplet magnon near the antiferromagnetic wave vector, $k \approx \pi$. Near zero wave vector the lowest energy excitation is a pair of magnons. We calculate the neutron-scattering cross section near $k = 0$, using a Landau-Ginzburg model and alternatively exact S-matrix results for the O(3) nonlinear σ model. The cross section is proportional to k^2. As a function of energy, it shows a rounded peak somewhat above the two-magnon threshold. The effects of anisotropy are also considered.

I. INTRODUCTION

One-dimensional Heisenberg antiferromagnets, with Hamiltonian

$$H = J \sum_i \vec{S}_i \cdot \vec{S}_{i+1}, \quad J > 0 \tag{1.1}$$

have disordered ground states. (Here the $\vec{S}_i$'s are quantum spin operators, of spin s.) As argued by Haldane,[1] there is an excitation gap for integer spin. Field-theory arguments predict that the gap is smallest at the antiferromagnetic wave vector, $k = \pi$. Here there is a triplet of magnons, with energy-momentum relation

$$E \cong \sqrt{\Delta^2 + v^2(k-\pi)^2}, \tag{1.2}$$

where Δ is the magnon gap and v is the spin-wave velocity (we adopt units in which the lattice spacing and $\hbar$ equal 1). On the other hand, near zero wave vector the lowest excitation is a pair of magnons (total wave vector $k \approx 2\pi \equiv 0$). This spectrum has been partially verified[2] by numerical simulations on chains of length up to 32. In particular, the gap at $k = 0$ appears to be very close to twice the gap at $k = \pi$. However, the two-particle nature of the excitations near 0 has not, to our knowledge, been tested. Nearly all reported experimental data on the dispersion relation from neutron scattering in $CsNiCl_3$,[3,4] $Ni(C_2H_8N_2)_2NO_2(ClO_4)$ (NENP),[5] and $RbNiCl_3$ (Ref. 6) have been near $k = \pi$. One exception is Ref. 4. In this paper, we report on detailed theoretical calculations of the neutron-scattering cross section near $k = 0$.

We begin with a couple of general observations about the form of this cross section. It is proportional to the spin-correlation function:

$$\mathcal{S}(k,\omega) \equiv \frac{1}{L}\sum_{a,b}\int dt\, e^{i\omega t - ik(a-b)} < 0|\vec{S}(a,t)\cdot\vec{S}(b,0)|0 > . \tag{1.3}$$

Here a and b are integers labeling points on the lattice; L is the length of the system. We set the temperature to zero throughout; this is not a serious limitation since experiments can be performed well below the gap ($\approx$15 K in NENP). Since the ground state $|0>$ is a spin singlet, $\sum_a \vec{S}(a,t)|0> = 0$, $\mathcal{S}(0,\omega) = 0$. For small k the correlation function is quadratic:

$$\mathcal{S}(k,\omega) \to k^2 \frac{1}{L}\sum_{a,b}\frac{(a-b)^2}{2}\int dt e^{i\omega t} < 0|\vec{S}(a,t)\cdot\vec{S}(b,0)|0 > . \tag{1.4}$$

We further note that assuming the two-magnon picture is correct, we should expect $\mathcal{S}(k,\omega)$ to vanish for $\omega < 2\Delta$. Detailed calculations, presented below, show an asymmetric rounded peak in $\mathcal{S}(k,\omega)$ as a function of ω, with maximum somewhat above 2Δ.

The field-theory treatment,[1] based on the large-s limit, introduces two fields $\vec{\phi}$ and $\vec{l}$ representing the staggered and uniform long-wavelength components of the spin operators ($\vec{S}_a$):

$$\vec{S}_a \approx s(-1)^a \vec{\phi}(a) + \vec{l}(a). \tag{1.5}$$

$\vec{\phi}$ and $\vec{l}$ are assumed to vary slowly on the lattice scale. (This is a fair approximation even for $s = 1$ where the correlation length is about seven lattice spacings.) The staggered and uniform magnetization do not commute, but rather obey

$$[l^i(x),\phi^j(y)] = i\epsilon^{ijk}\phi^k\delta(x-y), \quad (1.6)$$

$$[l^i(x),l^j(y)] = i\epsilon^{ijk}l^k\delta(x-y). \quad (1.7)$$

We make the basic semiclassical approximation (valid for large s and/or long wavelengths) that the components of $\vec{\phi}$ commute with each other. These commutation relations can be realized by writing $\vec{l} = \vec{\phi}\times\vec{\Pi}$, where $\vec{\Pi}$ is the canonical momentum variable conjugate to $\vec{\phi}$, $\vec{\Pi} \propto \partial\vec{\phi}/\partial t$. Since the staggered magnetization $\vec{\phi}$ is linear in magnon creation and annihilation operators, we see immediately that the uniform magnetization is a two-magnon operator. Expanding the Heisenberg Hamiltonian in the continuum fields gives the nonlinear σ model with Hamiltonian density

$$\mathcal{H} = \frac{v}{2}\left[g\vec{l}^2 + \frac{1}{g}\left(\frac{\partial\vec{\phi}}{\partial x}\right)^2\right], \quad (1.8)$$

where the spin-wave velocity is $v = 2Js$ and the coupling constant is $g = 2/s$. (The topological angle is zero for integer spin.) Thus the correlation function for k near 0 is given, in the continuum limit, by

$$\mathcal{S}(k,\omega) \approx \frac{1}{v^2g^2}\int dx\,dt\,e^{i(kx-\omega t)}\left\langle 0\left|\left(\vec{\phi}\times\frac{\partial\vec{\phi}}{\partial t}\right)(x,t)\cdot\left(\vec{\phi}\times\frac{\partial\vec{\phi}}{\partial t}\right)(0,0)\right|0\right\rangle. \quad (1.9)$$

The constraints on the length of the spin vectors translate into the nonlinear constraints on the σ-model fields: $\vec{\phi}^2 = 1$, $\vec{\phi}\cdot\vec{l} = 0$. Although the nonlinear σ model is highly nontrivial, much is known about it in one dimension, from the renormalization group, the large-n limit, and the exact S matrix. The spectrum consists of a triplet of massive magnons (with mass $\Delta \propto e^{-\pi s}$); fluctuation effects eliminate the constraint on the field $\vec{\phi}$, allowing it to have three degrees of freedom, instead of two. A much simpler model which has qualitatively similar behavior (and essentially arises in the large-n limit) is the Landau-Ginzburg model:

$$\mathcal{H} = \frac{v}{2}\vec{\Pi}^2 + \frac{v}{2}\left(\frac{\partial\vec{\phi}}{\partial x}\right)^2 + \frac{\Delta^2}{2v}\vec{\phi}^2 + \lambda\vec{\phi}^4. \quad (1.10)$$

Here the constraint on the field $\vec{\phi}$ is relaxed and the mass Δ is put in by hand. The coupling λ produces a repulsive interaction between the bosons. (The field $\vec{\phi}$ has been rescaled.) A simple mean-field theory is now obtained if we treat[7] the model perturbatively in λ. This has been used to treat a number of other properties of Haldane-gap antiferromagnets. In what follows, we first calculate the correlation function in the free-boson approximation, $\lambda = 0$. We then give the exact result for the σ model. They are qualitatively similar.

II. FREE-BOSON APPROXIMATION

We expand the staggered magnetization field $\vec{\phi}$ in magnon annihilation operators $\vec{a}_k$. It is convenient to denote the Lorentz-invariant contraction of two vectors by $\mathbf{a}\cdot\mathbf{b} \equiv a_0b_0 - a_1b_1$, and apply it to the space-time and energy-momentum two vectors: $\mathbf{X}_\mu = (\mathbf{X}_0,\mathbf{X}_1) \equiv (t, x/v)$ and $\mathbf{K}_\mu = (\mathbf{K}_0,\mathbf{K}_1) \equiv (\omega, vk)$. (The appearance of the dot product will always signify two vectors.) We adopt the relativistic normalization of the annihilation operators:

$$[a_k, a^\dagger_{k'}] = 4\pi v\omega_k\delta(k-k'), \quad (2.1)$$

where $\omega_k = \sqrt{\Delta^2 + (vk)^2}$. The mode expansion then takes the form

$$\vec{\phi}(x,t) = \int\frac{dk}{4\pi\omega_k}(e^{-i\mathbf{K}\cdot\mathbf{X}}\vec{a}_k + e^{i\mathbf{K}\cdot\mathbf{X}}\vec{a}^\dagger_k). \quad (2.2)$$

Here the dot product $\mathbf{K}\cdot\mathbf{X}$ represents the expression $(\omega_k t - kx)$. The uniform magnetization density, $\vec{l} = (1/v)\vec{\phi}\times\partial\vec{\phi}/\partial t$ contains four terms each with two-magnon annihilation or creation operators. To calculate the zero-temperature correlation function, we only need the term with two creation operators, and its Hermitian conjugate. The double-creation term is

$$l^3_c(x,t) = i\int\frac{dk'dk''}{16\pi^2 v\omega_{k'}\omega_{k''}}(\omega_{k''}-\omega_{k'})e^{i(\mathbf{K}'+\mathbf{K}'')\cdot\mathbf{X}}a^{1\dagger}_{k'}a^{2\dagger}_{k''}. \quad (2.3)$$

Note that the 3 component of the uniform magnetization involves the 1 and 2 components of the staggered magnetization. Thus we obtain

$$< 0|l^3(x,t)l^3(0,0)|0 >$$

$$= \int\frac{dk'dk''}{16\pi^2\omega_{k'}\omega_{k''}}e^{-i(\mathbf{K}'-\mathbf{K}'')\cdot\mathbf{X}}(\omega_{k''}-\omega_{k'})^2. \quad (2.4)$$

Fourier transforming, we obtain the 33 element of the correlation function in the form

$$\mathcal{S}^{33}(\mathbf{K}_\mu) = \int\frac{dk'dk''}{16\pi^2\omega_{k'}\omega_{k''}}(\omega_{k'}-\omega_{k''})^2(2\pi)^2 \times\delta^2(\mathbf{K}-\mathbf{K}'-\mathbf{K}''). \quad (2.5)$$

Here, $\delta^2(\mathbf{K})$ represents the Lorentz-invariant product, $\delta(\mathbf{K}_0)\delta(\mathbf{K}_1)$. Taking into account the Jacobian factor, we obtain

$$\mathcal{S}^{33}(\mathbf{K}_\mu) = \frac{(\mathbf{K}'_0 - \mathbf{K}''_0)^2}{2v^2|\mathbf{K}'_0\mathbf{K}''_1 - \mathbf{K}''_0\mathbf{K}'_1|}, \quad (2.6)$$

where $\mathbf{K}'$ and $\mathbf{K}''$ now represent one of the two solutions of the energy-momentum conservation equations. These are given by

$$\mathbf{K}'_{1\pm} = \left(vk \pm \frac{\omega\sqrt{\mathbf{K}\cdot\mathbf{K}-4\Delta^2}}{\sqrt{\mathbf{K}\cdot\mathbf{K}}} \right) \Big/ 2, \qquad (2.7)$$

$$\mathbf{K}'_{0\pm} = \left(\omega \pm \frac{vk\sqrt{\mathbf{K}\cdot\mathbf{K}-4\Delta^2}}{\sqrt{\mathbf{K}\cdot\mathbf{K}}} \right) \Big/ 2. \qquad (2.8)$$

Substituting the above expressions for the wave vectors and energies of the two magnons, we obtain

$$\mathcal{S}(\mathbf{K}_\mu) = \frac{3k^2\sqrt{\mathbf{K}\cdot\mathbf{K}-4\Delta^2}}{(\mathbf{K}\cdot\mathbf{K})^{3/2}}\theta(\mathbf{K}\cdot\mathbf{K}-4\Delta^2). \qquad (2.9)$$

(The factor of 3 arises from summing over the three components of l^a.) Several features of this expression are obvious. For fixed ω it vanishes quadratically as $k \to 0$, as expected from general principles. For fixed nonzero k, it vanishes for ω less than the threshold value, $\omega_{\rm th}(k) = \sqrt{v^2k^2+4\Delta^2}$, and rises as the square root of $\omega - \omega_{\rm th}$. It goes through a rounded maximum at:

$$\omega_{\rm max} \equiv \sqrt{6\Delta^2 + v^2k^2} \qquad (2.10)$$

of height $(k/\Delta)^2/2\sqrt{3}$ and then decreases as $1/\omega^2$ at large frequencies. Note that for small k, $\omega_{\rm th} \approx 2\Delta$ and $\omega_{\rm max} \approx \sqrt{6}\Delta \approx 2.5\Delta$.

III. NONLINEAR σ MODEL

By imposing requirements such as unitarity and crossing symmetry, an exact S matrix has been proposed[8] for the O(3) nonlinear σ model. The construction of this S matrix requires certain assumptions, basically that the spectrum is "minimal," i.e., it consists only of the triplet of massive bosons, with no bound states. The S matrix has been found by these methods for the O(n) model for all n and has been checked to O($1/n^2$) against the $1/n$ expansion of the σ model. This technique has been extended to calculate form factors.[9] The spin-correlation function can be expressed in terms of form factors by inserting a complete set of asymptotic states between the two factors of $\vec{l}$ in Eq. (1.9):

$$< 0|l^a(\mathbf{X}_\mu)l^a(0)|0> = \sum_n < 0|l^a(\mathbf{X}_\mu)|n>< n|l^a(0)|0> . \qquad (3.1)$$

It follows from Lorentz invariance that

$$< 0|l^a(\mathbf{X}_\mu)|n> = e^{i\mathbf{K}_n\cdot\mathbf{X}} < 0|l^a(0)|n> . \qquad (3.2)$$

Thus we obtain the correlation function

$$\mathcal{S}^{aa}(\mathbf{K}_\mu) = \sum_n |< 0|l^a(0)|n>|^2(2\pi)^2\delta^2(\mathbf{K}_n - \mathbf{K}). \qquad (3.3)$$

The lowest-energy intermediate state $|n>$ is the two-magnon state. This follows since the vacuum state $|0>$ is a singlet, so $< 0|l^a|0> = 0$ and the one-magnon state is odd under the discrete symmetry $\vec{\phi} \to -\vec{\phi}$, whereas $\vec{l}$ is even. The next-lowest-energy intermediate state is the four-magnon state. Thus in the range $4\Delta^2 < \mathbf{K}\cdot\mathbf{K} < 16\Delta^2$, only the two-magnon state contributes:

$$\mathcal{S}^{aa}(\mathbf{K}_\mu) = \int \frac{dk'dk''}{16\pi^2\omega_{k'}\omega_{k''}v^2} < 0|l^3(0)|1,k';2,k''>< 1,k';2,k''|l^3(0)|0> (2\pi)^2\delta^2(\mathbf{K}'+\mathbf{K}''-\mathbf{K}) \qquad (3.4)$$

(for $\mathbf{K}\cdot\mathbf{K} < 16\Delta^2$). [One-particle states are defined to have the normalization $|k> = a_k^\dagger|0>$ such that the resolution of the identity in the one-particle subspace of the Fock space is $I = \int dk|k>< k|/(4\pi\omega_k v)$.] Thus, in this frequency range, we only need the form factor: $< 0|l^3(0)|1,k';2,k''>$. Noting that $\vec{l}$ is the 0 component of the Lorentz two vector $\vec{J}_\mu \equiv (\vec{\phi}\times\partial_\mu\vec{\phi})/vg$, and that the form factor is odd under exchanging k' and k'', we see that

$$< 0|l^3(0)|1,k';2,k''> = i(\omega_{k''} - \omega_{k'})G(\theta), \qquad (3.5)$$

where G depends only on the Lorentz-invariant quantity $\mathbf{K}'\cdot\mathbf{K}'' = \frac{1}{2}\mathbf{K}\cdot\mathbf{K} - \Delta^2$. This is conveniently expressed in terms of rapidities, $\mathbf{K}'_\mu = \Delta(\cosh\theta',\sinh\theta')$, $\mathbf{K}''_\mu = \Delta(\cosh\theta'',\sinh\theta'')$, $\mathbf{K}'\cdot\mathbf{K}'' = \Delta^2\cosh(\theta)$, where $\theta \equiv \theta' - \theta''$. The function $G(\theta)$ may be pulled out of the integral in Eq. (3.4), leaving

$$\mathcal{S}^{33}(\mathbf{K}_\mu) = |G(\theta)|^2 \int \frac{dk'dk''}{16\pi^2\omega_{k'}\omega_{k''}}(\omega_{k''} - \omega_{k'})^2(2\pi)^2 \times\delta^2(\mathbf{K}'+\mathbf{K}''-\mathbf{K}). \qquad (3.6)$$

The integral is the same one encountered in the free-boson approximation Eq. (2.5), leaving

$$\mathcal{S}(\mathbf{K}_\mu) = |G(\theta)|^2\frac{3k^2\sqrt{\mathbf{K}\cdot\mathbf{K}-4\Delta^2}}{(\mathbf{K}\cdot\mathbf{K})^{3/2}} \quad (\text{for } 4\Delta^2 < \mathbf{K}\cdot\mathbf{K} < 16\Delta^2). \qquad (3.7)$$

The exact form factor of Karowski and Weisz[9] for the O(n) σ model gives

$$G_n(\theta) = \exp\left(2\int_0^\infty \frac{dx}{x}\frac{(e^{-2x/(n-2)}-1)\sin^2[x(i\pi-\theta)/2\pi]}{(e^x+1)\sinh(x)}\right). \qquad (3.8)$$

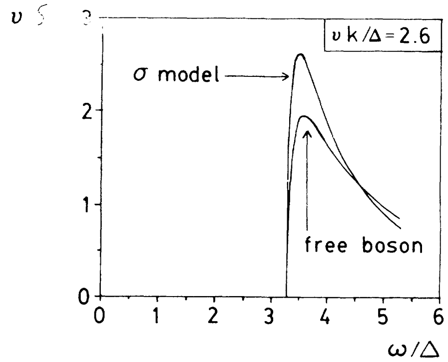

FIG. 1. $\mathcal{S}(\omega,k)$ for $k = 2.6\Delta/v$ from the free-boson and nonlinear σ models.

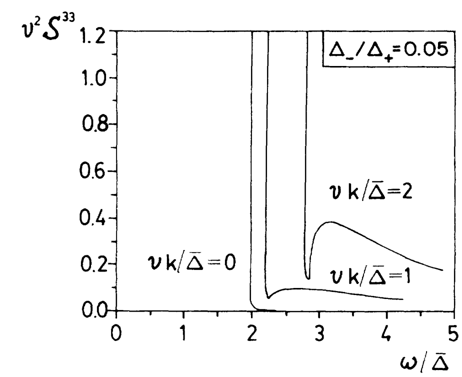

FIG. 3. $\mathcal{S}^{33}(\omega,k)$ for mass ratio $\Delta_-/\Delta_+ = 0.05$ for several values of k from the free-boson model.

Note that at $n \to \infty$ we obtain the free-boson result, $G(\theta) = 1$. Evaluating the integral for $n = 3$, gives

$$G(\theta) = \frac{\pi}{4}\frac{\Gamma(\frac{1}{2} - \frac{i\theta}{2\pi})\Gamma(\frac{3}{2} + \frac{i\theta}{2\pi})}{\Gamma(1 - \frac{i\theta}{2\pi})\Gamma(2 + \frac{i\theta}{2\pi})}, \tag{3.9}$$

where $\Gamma(z)$ is Euler's gamma function. Thus

$$|G(\theta)|^2 = \frac{\pi^4}{64}\frac{1 + (\theta/\pi)^2}{1 + (\theta/2\pi)^2}\left(\frac{\tanh\theta/2}{\theta/2}\right)^2. \tag{3.10}$$

At small θ this behaves as: $|G(\theta)|^2 \approx 1.52(1 - 0.09\theta^2)$. At large θ, $|G(\theta)|^2 \approx \pi^4/4\theta^2$. From the definition of θ we see that $\mathbf{K}\cdot\mathbf{K} = 4\Delta^2\cosh^2(\theta/2)$, so $\theta \to 0$ at the threshold: $\mathbf{K}\cdot\mathbf{K} - 4\Delta^2 \approx \Delta^2\theta^2$ and $\theta \to \infty$ at large energy: $\mathbf{K}\cdot\mathbf{K} \approx \Delta^2 e^\theta$. The free-boson and nonlinear-σ-model results are qualitatively similar. The effect of the interaction between the bosons is to narrow the peak and to raise the height at the maximum. (The free-boson and nonlinear-σ-model predictions are compared in Fig. 1.) Note that the normalization of the form factor is universal since, by crossing symmetry and translation invariance,

$$< 1,k|l^3(\mathbf{X}_\mu)|2,k >= 2i\omega_k G(i\pi). \tag{3.11}$$

Integrating over x, gives the 3 component of the total spin operator:

$$\int dx\, l^3(\mathbf{X}_\mu) = \sum_i S_i^3 \equiv S_T^3. \tag{3.12}$$

Since the single-magnon states may be decomposed into eigenstates of S_T^3, with eigenvalues ± 1, we obtain

$$\begin{aligned} < 1,k|l^3(\mathbf{X}_\mu)|2,k > &=< 1,k|S_T^3|2,k > /L \\ &= i < 1,k|1,k > /L = i2\omega_k. \end{aligned} \tag{3.13}$$

Hence, $G(i\pi) = 1$. Thus the overall scale of the correlation function is predicted by the σ model. The correlation function is plotted versus ω for various values of k in Fig. 2.

IV. EFFECTS OF ANISOTROPY

The most well-studied highly one-dimensional spin-1 antiferromagnet, NENP, contains significant anisotropy, assumed to be largely of crystal-field origin. This can be included in the nonlinear σ model by adding additional terms to the Hamiltonian density of the form

$$\delta\mathcal{H} = a(\phi^3)^2 + b(\phi^1)^2. \tag{4.1}$$

In the Landau-Ginzburg model, we simply allow for three different mass terms:

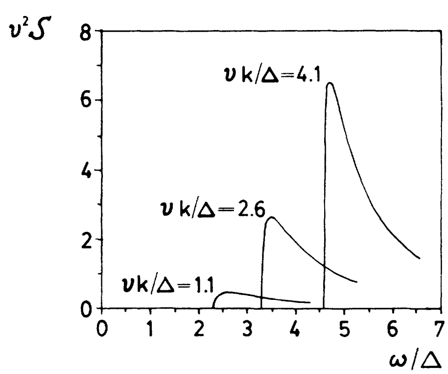

FIG. 2. $\mathcal{S}(\omega,k)$ for several values of k from the nonlinear σ model.

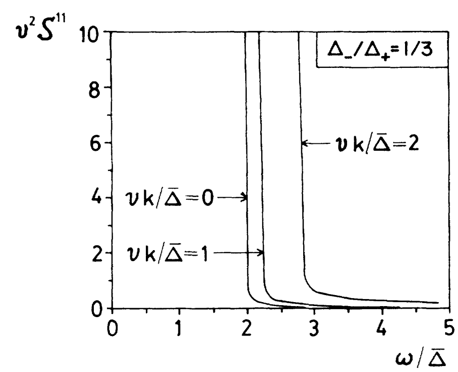

FIG. 4. $\mathcal{S}^{11}(\omega,k)$ for mass ratio $\Delta_-/\Delta_+ = \frac{1}{3}$ for several values of k from the free-boson model.

$$\mathcal{H} = \frac{v}{2}\vec{\Pi}^2 + \frac{v}{2}\left(\frac{\partial\vec{\phi}}{\partial x}\right)^2 + \sum_i \frac{\Delta_i^2}{2v}\phi_i^2 + \lambda\vec{\phi}^4. \tag{4.2}$$

Neutron-scattering experiments (near $k = \pi$) on NENP indicate that $\Delta_1 \approx 13$ K, $\Delta_2 \approx 15$ K, and $\Delta_3 \approx 30$ K.[5]

Since the 3 component of the uniform magnetization operator involves the one and two magnons, we expect that $\mathcal{S}^{33}$ will show a relatively small anisotropy while $\mathcal{S}^{11}$ and $\mathcal{S}^{22}$ will show a large one. Note that with anisotropy, the ground state is no longer a zero eigenstate of the total spin operator so that $\mathcal{S}(\omega, k)$ need no longer vanish at $k \to 0$. It can readily be seen that for small anisotropy, $\mathcal{S}(\omega, 0)$ is of quadratic order in the crystal-field term in the Hamiltonian (i.e., of quadratic order in the mass difference). The two-particle threshold in $\mathcal{S}^{33}$ now occurs at $\omega = \Delta_1 + \Delta_2$, etc.

We may readily repeat the free-boson calculation of $\mathcal{S}^{33}$ with anisotropy. Defining $\Delta_\pm \equiv \Delta_1 \pm \Delta_2$, we find

$$\mathcal{S}^{33}(\omega, k) = \left(\mathbf{K}_1^2\sqrt{[(\mathbf{K}\cdot\mathbf{K})^2 - \Delta_+^2][(\mathbf{K}\cdot\mathbf{K})^2 - \Delta_-^2]} + \frac{\Delta_+^2\Delta_-^2\mathbf{K}_0^2}{\sqrt{[(\mathbf{K}\cdot\mathbf{K})^2 - \Delta_+^2][(\mathbf{K}\cdot\mathbf{K})^2 - \Delta_-^2]}}\right)\frac{\theta(\mathbf{K}\cdot\mathbf{K} - \Delta_+^2)}{v^2(\mathbf{K}\cdot\mathbf{K})^2}. \tag{4.3}$$

As expected, $\mathcal{S}^{33}(\omega, k)$ no longer vanishes at $k \to 0$, but is rather of $O(\Delta_-^2)$. $\mathcal{S}^{33}(\omega, k)$ is now singular at the threshold due to the diverging density of states. For small anisotropy, there is a narrow peak near threshold and then a broader one at larger ω. The first peak is difficult to observe except at very small ω and k. For larger anisotropy $\mathcal{S}^{33}(\omega, k)$ is a monotonically decreasing function of ω. $\mathcal{S}^{33}(\omega, k)$ is shown in Fig. 3 for a small anistropy: $\Delta_-/\Delta_+ = 0.05$ and $\mathcal{S}^{11}(\omega, k)$ in Fig. 4 for a larger one: $\Delta_-/\Delta_+ = 0.33$, corresponding roughly to the situation in NENP. [$\bar{\Delta} \equiv (\Delta_1 + \Delta_2)/2$.] Anisotropy actually makes the $k \approx 0$ two-magnon peaks easier to observe since it makes them narrower and nonvanishing at $k \to 0$.

Exact results are not available for the nonlinear σ model with anistropy added, but based on our experience with the isotropic case, we might expect the results to be qualitatively similar to the free-boson approximation.

Neutron-scattering experiments on $CsNiCl_3$ (Ref. 4) seem to indicate a broader peak near $k \approx 0$ than near $k \approx \pi$, as predicted by the present theory. Experiments on NENP would probably be much more conclusive in this regard since they can be done at temperatures well below the gap, three-dimensional effects are smaller, and the significant anisotropy makes the signal easier to see, as mentioned above.

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