Dynamic Correlations in a Classical Two-dimensional Heisenberg Antiferromagnet

G. M. Wysin
Department of Physics, Cardwell Hall,
Kansas State University, Manhattan, KS 66506-2601
and
A. R. Bishop
Theoretical Division and Center for Nonlinear Studies
Los Alamos National Laboratory, Los Alamos, NM 87545

Abstract

A Monte Carlo–molecular dynamics calculation of the dynamic structure function $S(\mathbf{q},\omega)$ for the classical two-dimensional isotropic Heisenberg antiferromagnet is presented. For wavevectors near the antiferromagnetic Bragg point, $S(\mathbf{q},\omega)$ is well-described by a product of Lorentzians representing damped spin waves. For adequately low temperatures, the dependence of the spin wave frequency and damping on wavevector and temperature are consistent with a dynamic scaling description of Chakravarty, Halperin and Nelson. Even for higher temperatures a scaling description is quite well satisfied, but with a modified scaling frequency.

Phys. Rev. B 42, 810 (1990).

I. Introduction

Studies of spin dynamics in quasi-2-dimensional materials have received a significant boost in the last several years because of the convergence of: (a) improved quasi-2-dimensional magnetic materials; (b) low-frequency, long-wavelength inelastic neutron scattering data; and (c) simulation capabilities for large sized lattices. Materials with various spin symmetries have been studied, probing vortex, domain wall and spin wave dynamics and interactions.[1]

In this context, the copper oxide based high-temperature superconductors in their undoped (antiferromagnetic, non-superconducting) phase are now appreciated to be excellent examples of very 2-dimensional, antiferromagnetic, near-isotropic Heisenberg Hamiltonians.[2] This antiferromagnetism has been probed[2] by preliminary inelastic neutron scattering, NMR, etc. Comparisons with theory of the 2-D isotropic Heisenberg model are very successful, except for a small Dzyaloshinsky-Moriya term appearing in the real materials. This experimental-theoretical agreement is particularly well-confirmed for static spin correlations which are very long ranged, even at substantial temperatures. For dynamics, the data is much less complete at present, and theory[3] has emphasized a dynamic scaling expectation and its consequences. The real materials are extremely quantum (S=1/2) and the theory procedes by mapping the quantum isotropic Heisenberg Hamiltonian to a quantum O(3) nonlinear sigma model, whose scaling properties are studied by including quantum fluctuations into a classical O(3) model. Furthermore, the O(3) model can be mapped to a ferromagnetic rotor model at sufficiently low frequencies and long wavelengths, and this latter classical model has been studied numerically in a Langevin molecular dynamics (MD) simulation,[4] described below. This simulation appears to support the analytic dynamic scaling expectations. An alternative analytic procedure has been suggested by Reiter,[5] who used a low-T and (1/S) perturbative memory function approach for dynamic correlation functions. It is not clear whether this approach yields subtle violations of dynamic scaling, without further explicit evaluations.

It is of interest to explore the dynamics of the antiferromagnetic Heisenberg model directly in order to confirm the scaling behavior, to extend expectations (which can be

compared with experiment) outside the scaling regime, and to incorporate additional symmetry-breaking terms, inter-plane coupling, etc. The most complete approach is quantum Monte Carlo. This has been implemented for static properties,[6,7] and extracting frequency dependence from the imaginary time simulation data is probably also practical within some reasonable restrictions on the form of the dynamic structure function. Our aim in this paper is more limited. Namely, we wish to implement a classical ($S = \infty$) MD simulation to test dynamic scaling, and to establish a form for the scaling function and observe deviations at higher T, in the original discrete antiferromagnetic isotropic Heisenberg model. This avoids mapping to O(3) sigma or rotor models.

As mentioned above, Chakravarty, Halperin and Nelson (CHN) have developed a scaling theory[2] for the low temperature dynamic properties of the quantum Heisenberg antiferromagnet in two dimensions (QHAF). For adequately low temperatures, and wavevectors q close to the antiferromagnetic Bragg point, CHN have suggested that the dynamic properties of the QHAF can be obtained from dynamic properties of a *classical* lattice rotor model (CLRM). The CHN theory is a *dynamic scaling* theory, in the sense that dynamic properties, such as spin-wave frequencies, when measured at different temperatures, can be related to each other easily, provided that frequencies and wavelengths are appropriately re-scaled. The physical length scale at any temperature T is the spin-spin correlation length $\xi(T)$, which is the factor for re-scaling wavevectors.

For dynamics, the theory requires a temperature-dependent physical time scale $\tau_s(T)$, or alternatively a scaling frequency $\omega_s(T) = 2\pi/\tau_s$. The time-scale τ_s is the re-scaling factor for frequencies, and is closely related to the *damping rate* or linewidth for the spinwaves. The scaling assumption asserts that the frequency and wavevector dependencies of dynamic quantities will be determined only by the dimensionless re-scaled variables, $\mathbf{Q} = \mathbf{q}\xi$, and $\Omega = \omega\tau_s$, rather than by ω, q, and T separately. Through the use of a renormalization group (RG) analysis, CHN give specific expressions for the temperature dependence of the correlation length ξ and time τ_s.

To test the CHN theory, Tyc, Halperin, and Chakravarty (THC) have recently reported results[4] of a Langevin dynamics simulation of a classical lattice rotor model,

wherein the dynamic structure function, $S(\mathbf{q},\omega)$, was calculated numerically using a large lattice (256×256). For some set of wavevectors and temperatures satisfying the assumptions of the CHN theory, they obtained a reasonable fit to the scaling theory, by adjusting several parameters describing the scaling functions for spinwave frequency and damping rate.

It is our purpose here to report results of a related simulation, directly for the 2D *classical* Heisenberg antiferromagnet, which also substantially supports the dynamic scaling hypothesis. The classical isotropic spin model under consideration is

$$H = J \sum_{(\mathbf{n},\mathbf{m})} \vec{\mathbf{S}}_{\mathbf{n}} \cdot \vec{\mathbf{S}}_{\mathbf{m}} \tag{1.1}$$

where $J > 0$ is the exchange constant, $\vec{\mathbf{S}}_{\mathbf{n}}$ is a classical (three-component) spin vector at a lattice site $\mathbf{n}$ on a square lattice, and the sum is only over nearest neighbor pairs. Note that we use the symbol $\mathbf{q} = (1,1)\pi/a - \mathbf{k}$ to denote wavevectors measured from the antiferromagnetic Bragg point, $(1,1)\pi/a$. The lattice constant is a.

The principal quantity calculated in these simulations is the dynamic correlation function, $S(\mathbf{q},\omega)$, which is the space-time Fourier transform of the space and time displaced correlation function (see below). By fitting $S(\mathbf{q},\omega)$ to an assumed Lorentzian response function, the spin-wave frequency $\omega_{\mathbf{q}}$ and damping rate $\gamma_{\mathbf{q}}$ were determined numerically, and found to follow scaling relationships similar to those given by CHN. The scaling frequency $\omega_s(T)$ we obtained is consistent with the CHN theory except for temperatures greater than $T \approx 0.80$, which are probably outside of the scaling regime.

Our simulation differs from that of THC principally in three respects: i) THC have simulated a classical rotor model, with a Hamiltonian that includes a kinetic energy term in addition to a *ferromagnetic* nearest neighbor interaction. Of course, that choice was based on the CHN mapping to the QHAF mentioned above; to obtain properties of the $S = 1/2$ QHAF. Our simulation represents the opposite limit, $S \to \infty$; ii) By employing a Langevin type of equation to simulate finite temperatures, THC made use of the canonical ensemble, by coupling to a heat bath via random forces with a prescribed correlation, together with a small phenomenological damping. On the other hand, we have used a two-step simulation, using a (canonical ensemble) Monte Carlo

(MC) algorithm to produce states for a desired temperature, which were subsequently used as initial states for an energy-conserving spin-dynamics (MD) integration of the classical equations of motion, in a microcanonical ensemble. iii) We have used a 100 $\times$ 100 lattice ($L = 100$), smaller than that used by THC by a factor of 0.40 ($L = 256$). The lowest accessible temperature is determined roughly by the point at which the correlation length equals half the lattice size. The RG analysis and numerical results give $\xi \approx 130a$ at $T/JS^2 = 0.52$, and $\xi \approx 50a$ at $T/JS^2 = 0.57$, so the THC calculation is valid to only *slightly* lower temperatures than results presented here. The lowest accesible wavevector (either measured from $(0,0)\pi/a$ or from $(1,1)\pi/a$) is $q = 2\pi/La$, so that the major advantage of using a larger system size is access to much smaller wavevectors.

We begin in Sec. II by summarizing some general properties of the 2D Heisenberg antiferromagnet, within the scaling hypothesis. This includes a low precision fit to determine the correlation length for our finite system, followed by a description of the scaling hypothesis and scaling functions. In Sec. III and IV we will describe the details of the numerical simulation, along with a discussion of the fitting functions used for describing the spin-wave frequency and linewidths. We then present two methods in Sec. V for obtaining rough estimates of the scaling frequency $\omega_s(T)$, and from those obtain refined estimates. In the process of estimating $\omega_s(T)$, we graphically obtain scaling functions for spin-wave frequency and linewidth. Sec. VI contains concluding remarks.

II. Scaling in the Classical Heisenberg Antiferromagnet

A principal interest here is to describe the dynamic signature of the spinwaves in a 2D system. Of course, the lack of long range order at finite temperature, as given by the Mermin-Wagner theorem,[8] needs to be recognized. However, there *is* short range order, on length scales less than the correlation length. Therefore, the modification from 3D spinwave theory is that the spinwaves represent local perturbations from short range order, and can be used as well-defined modes as long as their wavelengths are much shorter than the correlation length(i.e., $q\xi >> 1$). In addition, the interactions among an equilibrium population of these modes at finite temperature leads to a damping of

the modes. This gives a mode at any wavevector q a finite lifetime, or equivalently, a nonzero frequency width $\gamma_{\mathbf{q}}$, about the center frequency, $\omega_{\mathbf{q}}$. For some range of wavevectors and temperatures, we are interested in finding $\gamma_{\mathbf{q}}$ and $\omega_{\mathbf{q}}$. For comparison, Tyč and Halperin,[9] and Becher and Reiter[10], have each given leading order perturbation expansions to obtain $\gamma_{\mathbf{q}}$ and $\omega_{\mathbf{q}}$, with somewhat different results, especially for the temperature dependence of $\gamma_{\mathbf{q}}$.

To determine the wavevector range where perturbation expansions will be applicable, we can make some estimates of the correlation length, and compare with other work. Shenker and Tobochnik[11] (SB) have determined the correlation length using a MC RG technique. Also, CHN have given an expression for the static structure function,

$$S(\mathbf{q}) = \frac{1}{3} < \vec{S}_{\mathbf{q}} \cdot \vec{S}_{-\mathbf{q}} > \tag{2.1}$$

where

$$\vec{S}_{\mathbf{q}} = \frac{1}{N} \sum_{\mathbf{n}} e^{i\mathbf{q}\cdot\mathbf{r}_{\mathbf{n}}} \vec{S}_{\mathbf{n}} \tag{2.2}$$

are the spatial Fourier modes, with N the number of spins in the system. The CHN RG analysis gives a modified Ornstein-Zernicke form (OZ),

$$S(\mathbf{q}) = S(q=0) \frac{1 + \frac{1}{2} B_f \ln[1 + (q\xi)^2]}{1 + (q\xi)^2}. \tag{2.3}$$

THC found $B_f \approx 4\pi/125$. The effect of the logarithm term is greatest at low temperature. If we take this expression to be valid, but restrict to small wavevectors, $q\xi \leq 1$, then the effect of the logarithm is negligible and the q-dependence is very closely of the OZ form. Then we can extract estimates of the correlation length from OZ plots as in Figure 1. The curves of $S(q=0)/S(q) - 1$ vs. q^2 for various temperatures have slopes that are the squares of the correlation length for that temperature. These data were produced from the MC calculation described below. Over the range of q shown, for temperatures $0.55 \leq T \leq 1.0$, the OZ form is fairly closely followed. We did not look carefully for the logarithmic corrections indicated above. The resulting correlation lengths are shown in Figure 2, and compared with the low temperature RG result of CHN,

$$\xi(T) = B_\xi a \frac{\exp(2\pi J S^2/T)}{1 + 2\pi J S^2/T}. \tag{2.4}$$

Here $B_\xi \approx 0.01$ is a fitting parameter that was obtained by Shenker and Tobochnik.[11] For lower temperatures, $T < 0.65$, the effects of the finite sized lattice clearly limit the correlation length. For temperatures greater than $T \approx 0.8$, our data also deviate from the low temperature RG prediction, but are consistent with the SB results and high temperature series.

For dynamic correlations, consider the temporal Fourier transform of the time-displaced correlation function,

$$S(\mathbf{q},\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dt\; e^{-i\omega t}\frac{1}{3} < S_{\mathbf{q}}(0)\cdot S_{-\mathbf{q}}(t) > . \tag{2.5}$$

According to the scaling theory of CHN, this dynamic structure function can be written in the scaled form,[2]

$$S(\mathbf{q},\omega) = \omega_s^{-1} S(q)\Phi(\mathbf{Q},\Omega), \tag{2.6}$$

where $\omega_s(T)$ is the temperature-dependent frequency scale ($= 2\pi/\tau_s$), and Φ is a dimensionless scaling function, of the scaled variables,

$$\mathbf{Q} \equiv \mathbf{q}\xi, \qquad \Omega \equiv \omega/\omega_s. \tag{2.7}$$

Note that $S(q)$ is actually assumed to be a function of $q\xi$ and T, by equation (2.3) above (but *not* of q itself). Then one can see that the essential feature of the scaling assumption is that the wavevector and frequency dependencies of $S(\mathbf{q},\omega)$ are assumed to be determined only by the scaled variables Q and Ω, independent of the temperature. The only effect of the temperature is a re-scaling of the magnitude of $S(\mathbf{q},\omega)$. CHN give the following expression for the frequency scale:

$$\omega_s = c\xi^{-1}(T/2\pi JS^2)^{1/2}, \tag{2.8}$$

with $c = \sqrt{8}JS^2a/\hbar$ being the zero temperature long wavelength spinwave velocity. In analyzing $S(\mathbf{q},\omega)$ data, we take $\omega_s(T)$ to be a freely adjustable function, determined via least squares curve fits to $S(\mathbf{q},\omega)$, as described below. The scaling function $\Phi(\mathbf{Q},\Omega)$ is also an unknown function, not prescribed by scaling theory, to be determined by the fits. However, this is intractable unless some simple functional form for Φ is assumed, possessing a limited number of fitting parameters.

This, to determine Φ, the data for $S(\mathbf{q},\omega)$ is to be fit to an assumed functional form, the simplest of which represents the frequency response of a damped harmonic oscillator. As one of the simplest possible choices for $\Phi(\mathbf{Q},\Omega)$, THC have used a sum of two Lorentzians, with equal widths but centered at opposite frequencies. However, when they compared their least squares curve fits with their original MD data for $S(\mathbf{q},\omega)$, at fixed $\mathbf{q}$, it was clear that the fitted curves fell below the data at low frequencies ($\omega < \omega_{\mathbf{q}}$), and fell consistently above the data at higher frequencies ($\omega > \omega_{\mathbf{q}}$). Our curve fits to a sum of Lorentzians for $S(\mathbf{q},\omega)$ also consistently give the same "anomolous" behavior for the temperatures presented here. This is apparently a consequence of the isotropic symmetry of the Heisenberg Hamiltonian, which places some restrictions on allowed forms for $S(\mathbf{q},\omega)$. To be specific, $\dot{S}(\mathbf{q},t \rightarrow 0)$ *must be zero.* This restriction can be shown to rule out a sum of Lorentzians as a choice for Φ; an alternative which dose not violate this requirement is a *product* of Lorentzians, and is shown below to be a natural choice.

We can demonstrate this restriction on $S(\mathbf{q},\omega)$ using the equations of motion as follows. The classical equations of motion resulting from Hamiltonian (1.1) are

$$\frac{d\vec{\mathbf{S}}_{\mathbf{n}}}{dt} = \vec{\mathbf{S}}_{\mathbf{n}} \times \vec{\mathbf{F}}_{\mathbf{n}}, \tag{2.9}$$

$$\vec{\mathbf{F}}_{\mathbf{n}} = -J \sum_{(\mathbf{n},\mathbf{m})} \vec{\mathbf{S}}_{\mathbf{m}}, \tag{2.10}$$

where $\vec{\mathbf{F}}_{\mathbf{n}}$ is the effective field acting on $\vec{\mathbf{S}}_{\mathbf{n}}$ produced by all of its nearest neighbors; the sum in Eqn. (2.10) is only over the nearest neighbors of $\mathbf{n}$.

Using the spatial Fourier modes defined above in equation (2.2), one component of the time-displaced correlation function can be written as

$$S^{\alpha\alpha}(\mathbf{q},t) = \frac{1}{N^2} \sum_{\mathbf{n},\mathbf{m}} e^{i\mathbf{q}\cdot(\mathbf{r_n}-\mathbf{r_m})} < S^{\alpha}_{\mathbf{n}}(0) S^{\alpha}_{\mathbf{m}}(t) > . \tag{2.11}$$

The $t = 0$ time derivative of this is

$$\dot{S}^{\alpha\alpha}(\mathbf{q},0) = < S^{\alpha}_{\mathbf{q}}(0) \dot{S}^{\alpha}_{-\mathbf{q}}(0) > = \frac{1}{N^2} \sum_{\mathbf{n},\mathbf{m}} e^{i\mathbf{q}\cdot(\mathbf{r_n}-\mathbf{r_m})} < S^{\alpha}_{\mathbf{n}}(0) \dot{S}^{\alpha}_{\mathbf{m}}(0) > . \tag{2.12}$$

This can be evaluated in terms of the equal-time averages, $< S^{\alpha}_{\mathbf{n}}(0)\dot{S}^{\alpha}_{\mathbf{m}}(0) >$, which are equilibrium averages that are readily evaluated. For example, consider $\alpha = x$, with equation of motion for this component

$$\dot{S}^{x}_{\mathbf{m}} = -J \sum_{(\mathbf{m},\mathbf{m}')} (S^{y}_{\mathbf{m}} S^{z}_{\mathbf{m}'} - S^{z}_{\mathbf{m}} S^{y}_{\mathbf{m}'}). \tag{2.13}$$

The sum is over the neighbors $\mathbf{m}'$ of site $\mathbf{m}$. This is multiplied by $S^{x}_{\mathbf{n}}$ and then the equilibrium expectation value is taken:

$$< S^{x}_{\mathbf{n}} \dot{S}^{x}_{\mathbf{m}} >= -J \sum_{(\mathbf{m},\mathbf{m}')} (< S^{x}_{\mathbf{n}} S^{y}_{\mathbf{m}} S^{z}_{\mathbf{m}'} > - < S^{x}_{\mathbf{n}} S^{z}_{\mathbf{m}} S^{y}_{\mathbf{m}'} >). \tag{2.14}$$

However, the isotropic Hamiltonian is invariant under a re-labeling of the y and z spin axes, so that $< S^{x}_{\mathbf{n}} S^{y}_{\mathbf{m}} S^{z}_{\mathbf{m}'} >=< S^{x}_{\mathbf{n}} S^{z}_{\mathbf{m}} S^{y}_{\mathbf{m}'} >$. Then in this case the two terms in Eq. (2.14) are identical and cancel, regardless of whether $\mathbf{n} = \mathbf{m}$, or whether $\mathbf{n}$ is a neighbor of $\mathbf{m}$ or not. Thus we conclude that for the Heisenberg model,

$$< S^{z}_{\mathbf{n}} \dot{S}^{z}_{\mathbf{m}} >=< S^{y}_{\mathbf{n}} \dot{S}^{y}_{\mathbf{m}} >=< S^{x}_{\mathbf{n}} \dot{S}^{z}_{\mathbf{m}} >= 0, \tag{2.15}$$

for *any* sites $\mathbf{n}$ and $\mathbf{m}$. For $\mathbf{n} = \mathbf{m}$ this is just a statement of the conserved spin length. However, since the isotropic symmetry makes it true more generally, then we also have

$$< S^{\alpha}_{\mathbf{q}}(0) \dot{S}^{\alpha}_{-\mathbf{q}}(0) >= 0, \tag{2.16}$$

for $\alpha = x, y$, or z (which are all equal). In similar fashion, for an XY-model, the invariance of the Hamiltonian with respect to re-labeling x and y spin axes, implies only that $< S^{z}_{\mathbf{q}}(0) \dot{S}^{z}_{-\mathbf{q}}(0) >$ is zero.

An apropriate form for $S(\mathbf{q}, t)$, consistent with the above restriction, is

$$S(\mathbf{q}, t) = A e^{-\gamma_{\mathbf{q}}|t|} \{\cos \omega_{\mathbf{q}} t + \frac{\gamma_{\mathbf{q}}}{\omega_{\mathbf{q}}} \mathrm{sign}(t) \sin \omega_{\mathbf{q}} t\}. \tag{2.17}$$

The parameters $\omega_{\mathbf{q}}$ and $\gamma_{\mathbf{q}}$ are to be fit as functions of $\mathbf{q}$ and T. This function is the response of an underdamped harmonic oscillator that starts with zero velocity from some initial displacement A, and is constructed to be an even real function of t. The

initial displacement A is identified as $S(\mathbf{q}, t = 0)$. The two-sided Fourier transform is also real;

$$S(\mathbf{q},\omega) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} dt\ S(\mathbf{q},t)e^{-i\omega t} = \frac{2A\gamma_\mathbf{q}(\gamma_\mathbf{q}^2+\omega_\mathbf{q}^2)/\pi}{[(\omega+\omega_\mathbf{q})^2+\gamma_\mathbf{q}^2][(\omega-\omega_\mathbf{q})^2+\gamma_\mathbf{q}^2]}, \tag{2.18}$$

and is a *product* of two symmetrically located Lorentzians.[12]

Alternatively, note that if a *sum* of symmetrically located Lorentzians is assumed for $S(\mathbf{q},\omega)$, i. e.

$$S(\mathbf{q},\omega) = \frac{A\gamma_\mathbf{q}}{2\pi}\{\frac{1}{(\omega+\omega_\mathbf{q})^2+\gamma_\mathbf{q}^2} + \frac{1}{(\omega-\omega)_\mathbf{q}^2+\gamma_\mathbf{q}^2}\}, \tag{2.19}$$

then the time-correlation is

$$S(\mathbf{q},t) = Ae^{-\gamma_\mathbf{q}|t|}\cos\omega_\mathbf{q}t. \tag{2.20}$$

This function has a discontinuous non-zero time derivative at $t = 0$; $\dot{S}(\mathbf{q},0^+) = -A\gamma_\mathbf{q}$, and $\dot{S}(\mathbf{q},0^-) = +A\gamma_\mathbf{q}$, and making it unsuitable for use in the present context, especially for strong damping, at short times (high frequencies).

There is also the possibility of fitting to the response of an *overdamped* harmonic oscillator, which may be relevant especially for higher temperatures. Then we would have a non-oscillatory time dependence,

$$S(\mathbf{q},t) = Ae^{-\gamma_\mathbf{q}|t|}\{\cosh\omega_\mathbf{q}t + \frac{\gamma_\mathbf{q}}{\omega_\mathbf{q}}\mathrm{sign}(t)\sinh\omega_\mathbf{q}t\}. \tag{2.21}$$

In this case the Fourier transform is also a product of Lorentzians, each centered at zero frequency, but having different widths:

$$S(\mathbf{q},\omega) = \frac{2A\gamma_\mathbf{q}(\gamma_\mathbf{q}^2-\omega_\mathbf{q}^2)/\pi}{[\omega^2+(\gamma_\mathbf{q}+\omega_\mathbf{q})^2][\omega^2+(\gamma_\mathbf{q}-\omega_\mathbf{q})^2]}\quad , \tag{2.22}$$

Generally, however, it was not possible to describe the behavior of $S(\mathbf{q},t)$ by these overdamped functions; in all cases discussed here the response of $S(\mathbf{q},t)$ involved oscillatory behavior, and was quite well described by the underdamped oscillator formulas, even for higher temperatures and $\mathbf{q}$ near $(1,1)\pi/a$.

To complete the scaling description, note that Eq. (2.18) has exactly the scaling form of Eq. (2.6), provided that the spinwave frequency and damping can be written in terms of undetermined, dimensionless scaled functions $\Omega(\mathbf{q}\xi)$ and $\Gamma(\mathbf{q}\xi)$:

$$\omega_{\mathbf{q}}(T) = \omega_s(T)\Omega(\mathbf{q}\xi), \tag{2.23}$$

$$\gamma_{\mathbf{q}}(T) = \omega_s(T)\Gamma(\mathbf{q}\xi). \tag{2.24}$$

All of the explicit temperature dependence of $\omega_{\mathbf{q}}$ and $\gamma_{\mathbf{q}}$ is given through the ω_s dependence. Then the assumed form of $\Phi(\mathbf{Q},\Omega)$ is (with $\mathbf{Q} = \mathbf{q}\xi$)

$$\Phi(\mathbf{Q},\Omega) = \frac{2\Gamma_{\mathbf{Q}}(\Gamma_{\mathbf{Q}}^2 + \Omega_{\mathbf{Q}}^2)/\pi}{[(\Omega+\Omega_{\mathbf{Q}})^2 + \Gamma_{\mathbf{Q}}^2][(\Omega-\Omega_{\mathbf{Q}})^2 + \Gamma_{\mathbf{Q}}^2]}. \tag{2.25}$$

This function is normalized such that

$$\int_{-\infty}^{+\infty} d\Omega\ \Phi(\mathbf{Q},\Omega) = 1, \tag{2.26}$$

and

$$\Phi(0,0) = \frac{2}{\pi\Gamma_0}, \tag{2.27}$$

with the assumption $\Omega_0 = 0$. It is interesting to note that Eq. (2.27) also would hold had we assumed a single Lorentzian for $\mathbf{Q} = 0$.

Equation (2.24) gives a direct method of estimating *relative* values of ω_s from MD data. The $q = 0$ spinwave linewidth directly gives $\omega_s\Gamma_0$. Of course this method requires first making the fit to obtain $\gamma_{\mathbf{q}}$. An alternative procedure is to make use of the $q = 0, \omega = 0$ datum, for which the scaling assumptions (2.6) and (2.27) above imply

$$\Gamma_0\omega_s = \frac{2}{\pi}\frac{S(0,t=0)}{S(0,\omega=0)}. \tag{2.28}$$

The advantage of this formula is that a fit is not required, although there is still an implicit assumption about the form of $\Phi(\mathbf{Q},\Omega)$ (as in Eq. (2.25)). However, Eq. (2.28) has a fairly intuitive interpretation, especially in terms of the time scale τ_s that results from it. Eq. (2.28) gives

$$\tau_s = \frac{\pi\Gamma_0}{2}\frac{1}{S(0,t=0)}\int_{-\infty}^{+\infty} dt\ S(0,t). \tag{2.29}$$

Apart from the numerical factors, τ_s is the ratio of the area under the $S(0,t)$ curve to the initial value $S(0,t=0)$, which is the simplest way to extract a physical time scale from these quantities. We used both of the above methods for estimating $\omega_s\Gamma_0$ from the numerical simulation data, obtaining very similar results. Both of these methods only give the product, $\omega_s\Gamma_0$, and Γ_0 is undetermined by this fitting procedure. For further analysis, we set $\Gamma_0 = 1$.

III. Numerical Simulation

Numerically we have simulated the classical spin dynamics of the isotropic Hamiltonian in equation (1.1). A 100 x 100 square lattice with periodic boundary conditions was used. The product JS^2 sets the energy scale for this model. Therefore, in numerical results presented here, temperatures will be measured in units of JS^2, frequencies are measured in units of $JS/\hbar$, and wavevectors are measured in units of $1/a$, where a is the lattice spacing.

The method of simulation is a combined Monte Carlo – molecular dynamics technique.[13] First a Monte Carlo simulation is used to equilibrate the system (in a canonical ensemble) to a desired temperature T. A standard Metropolis importance sampling scheme is used. The final state of the Monte Carlo calculation is then used as the initial configuration for an energy-conserving spin-dynamics simulation (in a microcanonical ensemble). Desired time-dependent correlation functions are obtained directly from the spin configuration as it evolves in time. The ensemble average of any time-dependent quantity is then obtained by repeating the spin dynamics calculation using additional initial configurations obtained from new Monte Carlo simulations, and averaging results over the separate initial configurations. Data presented here represent averages over 5 initial configurations.

In the Monte Carlo simulation, the initial state is one in which each spin is given a random initial direction (uniformly distributed on a unit sphere). To encourage the system to sample the allowed phase space in an unbiased manner, trial states for individual spins are also chosen randomly in direction, and then the acceptance rate is determined by the temperature. For temperatures higher than about $T/JS^2 > 0.4$ the resulting acceptance rate is greater than 15 % . For equilibration, the temperature is brought