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A Theory of Spin Dynamics in the Classical Two-Dimensional Heisenberg Magnet

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Abstract

The wave vector-dependent spin autocorrelation function of a classical Heisenberg model on a square lattice is calculated from the coupled-mode theory of spin dynamics. This theory is consistent with the spherical model for static spin correlations; as the temperature, T, approaches zero the inverse correlation length $\kappa \sim \exp(-\text{const.}/T)$. For a ferromagnetic exchange coupling, the decay rate of long wavelength fluctuations, $\Gamma(q)$, is proportional to $q^2T^{1/2}$ in the limit $(q/\kappa) \rightarrow \infty$, whereas in the opposite, hydrodynamic limit $\Gamma(q) \propto q^2 \{T \ln(\kappa/q)\}^{1/2}$. At the wave vector for incipient antiferromagnetic ordering, the decay rate is proportional to $\kappa T^{1/2}$, while the corresponding decay rate near the Brillouin zone centre is proportional to $(q^2T^{1/2}/\kappa)$. The coupled-mode equations for ferromagnetically and antiferromagnetically coupled models are solved numerically on a fine grid of wave vectors. The spin autocorrelation function, and its power spectrum, are surveyed over a wide range of temperatures and wave vectors.

1. Introduction

Magnetism in two dimensions is known to possess a variety of subtle features. Perhaps the best known, and the most firmly established, example is that the isotropic Heisenberg model does not support magnetic long range order at a finite temperature (Mermin and Wagner, 1966) whereas the Ising model orders at a finite temperature. However, it has been conjectured that the Heisenberg model at a finite temperature has a phase transition for which the susceptibility is infinite. To date, the type of transition in question, where the signature is an infinite correlation length without magnetic ordering, has only been established for an anisotropic version of the model (Berezisnkii, 1971; Kosterlitz and Thouless, 1973). These and other features of magnetism in two dimensions are reviewed by Mattis (1985).

Here, we address the nature of time-dependent spin correlations in the classical Heisenberg model on a square lattice. In our calculations, the equilibrium spin correlations are described by the spherical model. This estimate of the correlations is consistent with the Mermin-Wagner theorem (1966) since the spherical model has no phase transition for spatial dimension ≤ 2. For two dimensions, as the temperature, T, approaches zero the correlation length is proportional to exp(const./T).

We describe time-dependent spin correlation functions by the so-called coupled–mode theory of critical and paramagnetic fluctuations. For three dimensional magnets, this theory is unrivalled in its quantitative value (Cuccoli et al., 1989).

In subsequent sections, the spin autocorrelation function $F(\mathbf{q},t)$ is calculated for the full range of wave vectors in the Brillouin zone. Temperatures span the range $T = \infty$ down to a value where the correlation length is approximately 30 times the lattice spacing. Both ferromagnetic and antiferromagnetic exchange models are treated. In addition to $F(\mathbf{q},t)$, equal attention is given to its power spectrum $S(\mathbf{q},\omega)$ which is proportional to the signal monitored by inelastic neutron scattering from a

simple magnetic material (Collins, 1989). (As of now, no magnetic material has been discovered which is well described by the isotropic two dimensional Heisenberg model. The quasi-two dimensional magnets that have been studied possess significant magnetic anisotropy, which has a pronounced influence on the observed static and dynamic spin response functions.)

The Heisenberg magnet, spherical model, and coupled-mode theory are introduced in §§2,3. Numerical results for $F(\mathbf{q},t)$ and $S(\mathbf{q},\omega)$ are reviewed in §4. Estimates of the decay rates given in §5 characterize the very low temperature spin dynamics. Our findings are discussed in §6.

2. Model

Unit vector spins, $\{\mathbf{S}_a\}$, are arranged at sites labelled by the index a on a square lattice with a unit length a_o. Nearest neighbour spins interact through a Heisenberg exchange interaction of strength J. Hence, the Hamiltonian of our classical two-dimensional model is,

$$\mathcal{H} = \pm \tfrac{1}{2} J \sum_{a,b}^{n.n.} \mathbf{S}_a \cdot \mathbf{S}_b \,. \tag{2.1}$$

Here, the sum is restricted to all pairs of nearest neighbours. The upper and lower signs in (2.1) will be referred to as antiferromagnetic and ferromagnetic exchange couplings, respectively.

The theory utilized to describe the dynamic properties of (2.1) is consistent with the spherical model of equilibrium spin correlations. In this model, the isothermal susceptibility, $\chi(\mathbf{q})$, is expressed in terms of a dimensionless parameter, μ, which is related to the temperature, $T(k_B = \hbar = 1)$. Let us define,

$$\gamma_{\mathbf{q}} = \tfrac{1}{2}(\cos a_o q_x + \cos a_o q_y) \,. \tag{2.2}$$

The spherical model susceptibility is,

$$\chi(\mathbf{q}) = \{4J(\mu \pm \gamma_{\mathbf{q}})\}^{-1}, \tag{2.3}$$

and μ satisfies the transcendental equation,

$$(2\pi\mu\, J / 3T) = K(1/\mu), \tag{2.4}$$

where $K(x)$ is the complete elliptic integral. Fig. (1) shows graphically the relation between μ and a reduced temperature variable $\theta = (3T/4J)$. As θ approaches zero, μ tends to unity from above, and for a sufficiently small θ, which is found to be $\theta < 0.6$,

$$(\mu - 1) \sim 8 \exp(-\pi / \theta). \tag{2.5}$$

At low temperatures, the susceptibility is peaked at $\mathbf{q} = 0(\mathbf{w} = \pi(1,1)/a_o)$ for a ferromagnetic (antiferromagnetic) exchange coupling. Near these special wave vectors one obtains an Ornstein-Zernike expression for the susceptibility,

$$\chi(q) = \{Ja_o^2\,(\kappa^2 + q^2)\}^{-1}, \tag{2.6}$$

in which the inverse correlation length, κ, satisfies,

$$a_o\kappa = 2\ (\mu - 1)^{1/2} \sim 4\sqrt{2}\, \exp(-\pi / 2\theta);\ \theta \to 0. \tag{2.7}$$

Representative values of θ and κ as a function of μ are gathered in Table 1.

3. Coupled-mode Theory

The coupled-mode theory of critical and paramagnetic spin fluctuations is reviewed in several articles, e.g. Lovesey (1986) and Cuccoli et al. (1989). In view of

this, the following account is very brief, and aims to do no more than define our notation and essential equations.

Our spin autocorrelation function, $F(\mathbf{q},t)$, is normalized to the value one at time $t = 0$ for all values of the wave vector, $\mathbf{q}$. Denoting the thermal average of variables by angular brackets,

$$F(\mathbf{q},t) = < \mathbf{S}(\mathbf{q},t) \cdot \mathbf{S}(\mathbf{q},0) > /3T\chi(\mathbf{q}), \tag{3.1}$$

where $\mathbf{S}(\mathbf{q},t)$ is a spatial Fourier component of the spin density, and $\chi(\mathbf{q})$ is the isothermal susceptibility introduced in the previous section.

Coupled-mode theory can be viewed as a closure approximation to the infinite hierarchy of equations of motion for spin variables in the model (2.1). The corresponding set of equations for $F(\mathbf{q},t)$ are conveniently expressed in terms of a memory function, $K(\mathbf{q},t)$, which is defined through the equation,

$$\partial_t F(\mathbf{q},t) = -\int_0^t dt' \, F(\mathbf{q},t') \, K(\mathbf{q},t-t'). \tag{3.2}$$

In this format, closure is expressed as an approximation to $K(\mathbf{q},t)$ in terms of a spatial convolution of the product of two spin autocorrelation functions. A useful form of the expression for $K(\mathbf{q},t)$ is,

$$K(\mathbf{q},t) = \{T(4J)^2 / \chi(\mathbf{q})\} (\tfrac{1}{N}) \sum_{\mathbf{k}} (\gamma_{\mathbf{k}} - \gamma_{\mathbf{q}-\mathbf{k}})^2 \, \chi(\mathbf{k})\chi(\mathbf{q}-\mathbf{k}) \, F(\mathbf{k},t) \, F(\mathbf{q}-\mathbf{k},t) \, . \tag{3.3}$$

Here, the susceptibility is defined by (2.3), and N is the number of spins on the square lattice.

An expansion of $F(\mathbf{q},t)$ in powers of t begin with the terms,

$$F(\mathbf{q},t) = 1 - \tfrac{1}{2}(t\omega_o)^2 + \cdots . \tag{3.4}$$

From the defining equation (3.2) it follows that, the second frequency moment, $\omega_o^2(\mathbf{q})$, in (3.4) is the initial value of the memory function, viz.,

$$K(\mathbf{q},0) = \omega_0^2(\mathbf{q}). \tag{3.5}$$

The expression (3.3) provides the estimate,

$$\omega_0^2(\mathbf{q}) = 2T(1-\gamma_{\mathbf{q}})(\mu I(\mu) - 1)/\chi(\mathbf{q}), \tag{3.6}$$

in which $I(\mu)$ is the extended Watson integral. For the square lattice, $I(\mu)$ can be expressed in terms of the complete elliptic integral, encountered already in §2,

$$I(\mu) = 2K(1/\mu)/\pi\mu . \tag{3.7}$$

4. Numerical results

Numerical results for $F(\mathbf{q},t)$ have been obtained from (3.2) and (3.3) using a method described by Cuccoli et al. (1989). In the present case, the Brillouin zone is spanned by a mesh of equal square elements with a side of length $(\pi/24a_o)$. Results for $F(\mathbf{q},t)$ are given at three wave vectors denoted by $\mathbf{q}_1$, $\mathbf{q}_2$ and $\mathbf{q}_3$; in units of $(\pi/24a_o)$ these vectors are $\mathbf{q}_1 = (1,1)$, $\mathbf{q}_2 = (12,12)$ and $\mathbf{q}_3 = (24,24)$. Note that, $\mathbf{q}_3$ is the ferromagnetic zone boundary, and also the incipient antiferromagnetic ordering wave vector, i.e. $\mathbf{q}_3 = \mathbf{w}$ and $\gamma_{\mathbf{w}} = -1$. The wave vector $\mathbf{q}_2 = \tfrac{1}{2}\mathbf{w}$ is the antiferromagnetic zone boundary at which $\gamma_{\mathbf{q}} = 0$. Perhaps it is useful to observe that, in the limit of very low temperatures the second frequency moment, $\omega_o^2(\mathbf{q})$, given in (3.6) is proportional

to the square of the spin-wave frequency, i.e. $\omega_o^2\ \alpha(1-\gamma_{\mathbf{q}})^2$ and $\omega_o^2\ \alpha(1-\gamma_{\mathbf{q}}^2)$ for ferromagnetic and antiferromagnetic exchange couplings, respectively. This result leads us to expect that at low temperatures the response function,

$$S(\mathbf{q},\omega) = (1/2\pi) \int_{-\infty}^{\infty} dt\ \exp(-i\omega t)\ F(\mathbf{q},t), \tag{4.1}$$

is very narrow for small $\mathbf{q}$, e.g. $\mathbf{q}_1$. For antiferromagnetic coupling, $S(\mathbf{q},\omega)$ will also be narrow at $\mathbf{q}_3$, and relatively broad at $\mathbf{q}_2$. On the other hand, for ferromagnetic coupling the response function will broaden steadily by increasing $\mathbf{q}$ from zero through to $\mathbf{q}_3$.

At infinite temperature the properties of the Heisenberg model are independent of the sign of the exchange interaction, i.e. they are the same for ferromagnetic and antiferromagnetic interactions. Hence, the infinite temperature limit provides a sensible starting point for a study of dynamic spin fluctuations in the two model systems. Results for $F(\mathbf{q},t)$ and $S(\mathbf{q},\omega)$ for $T = \infty$ are shown in figs. (2) and (3). Looking at $F(\mathbf{q},t)$, the decay rate increases with increasing q and oscillations occur for the two largest wave vectors. Therefore, the corresponding response functions differ significantly from a gaussian function of ω, which might be one's first guess for the shape of the response function at infinite temperature.

Figs. (2) and (3) also contain results for $F(\mathbf{q},t)$ and $S(\mathbf{q},\omega)$, respectively, at low temperatures, cf. Table 1. The dramatic changes in $S(\mathbf{q}_3,\omega)$ with decreasing temperature are a signature of incipient antiferromagnetic ordering. Looking at the corresponding values of $F(\mathbf{q}_3,t)$ in fig. (2c), the slowing down of spin fluctuations with decreasing temperature is readily apparent. By comparison with what is found at $\mathbf{q}_3 = \mathbf{w}$ as a function of temperature, the fluctuations at $\mathbf{q}_2 = \frac{1}{2}\,\mathbf{w}$ are relatively benign functions of the temperature. The "squaring up" of $S(\mathbf{q}_2,\omega)$ with decreasing temperature might be interpreted as a premonition of a collective spin oscillation (spin-wave) in the ordered state. Turning attention to the smallest wave vector, $\mathbf{q}_1$, the

decay rate increases with decreasing temperature, which is the opposite of the trend at $\mathbf{q}_1$ for a ferromagnetically coupled system.

Data for a ferromagnetic exchange coupling are displayed in figs. (4) and (5). Slowing down of the spin fluctuations with decreasing temperature is quite apparent at $\mathbf{q}_1$. However, the changes in $F(\mathbf{q},t)$ and $S(\mathbf{q},\omega)$ at all three values of $\mathbf{q}$ brought about by decreasing the temperature from $\mu = 1.1$ ($\theta = 0.744$) to $\mu = 1.01$ ($\theta = 0.472$) are quite modest. Results for $\mu = \infty$ ($\theta = \infty$) are included for comparison.

At very low temperatures, such that $a_o \kappa << 1$, there is a dramatic slowing down of spin fluctuations at the wave vector which defines the incipient ordering. The decay rates at these special wave vectors are obtained from coupled-mode theory by an analysis which is outlined in the next section.

5. Decay rates

For a ferromagnetic coupling, the numerical results display the anticipated slowing down of long wavelength fluctuations as the temperature is decreased. This process in the autocorrelation function can be characterized by a decay rate, $\Gamma(q)$, which we will now estimate.

In the limit of low temperature and a small q equation (3.3) for the memory function approaches the result,

$$K(q,t) = (2JT(\kappa^2 + q^2)\, q^2\, a_o^2)\, \frac{1}{N} \sum_{\mathbf{k}} F(\mathbf{k},t)\, F(\mathbf{q}-\mathbf{k},t) / (\kappa^2 + k^2)\,. \qquad (5.1)$$

Here, the inverse correlation length, κ, is determined as a function of temperature by the relation (2.7). When the integral on the right-hand side of (5.1) is dominated by the slow processes at long wavelengths, the associated decay rate satisfies the integral equation,

$$\Gamma(q) = A_o q^2 \, (q^2 + \kappa^2) \int_q k dk \, / \{ \Gamma(k)(\kappa^2 + k^2) \}, \qquad (5.2)$$

where A_o is proportional to the temperature. For a square lattice,

$$A_o = (JTa_o^4 / 2\pi). \qquad (5.3)$$

One finds the following limiting forms for the decay rate:

(a) $(\kappa/q) \rightarrow \infty$; the hydrodynamic limit,

$$\Gamma(q) \sim q^2 \, \{2A_o \ln (\kappa / q)\}^{1/2}, \qquad (5.4)$$

and,

(b) $(\kappa/q) \rightarrow 0$; the critical limit,

$$\Gamma(q) \sim q^2 \, (A_o / 2)^{1/2}. \qquad (5.5)$$

As might be expected, in the critical limit the decay rate does not depend explicitly on the inverse correlation length. The results (5.4) and (5.5) are limiting cases of the general solution to (5.2). For the latter, it is prudent to introduce a dimensionless variable $\zeta = (q/\kappa)^2$. We find,

$$\Gamma(q) = q^2 \, (1 + \zeta) \{A_o \, [\ln (1 + 1/\zeta) - 1/(1 + \zeta)]\}^{1/2}, \qquad (5.6)$$

in which A_o is defined by (5.3).

The corresponding analysis for the antiferromagnetically coupled model is slightly more complicated. One needs equations for the decay rates at the zone centre, Γ_o, and at the ordering wave vector, Γ. The necessary equations for $\Gamma(q)$ and $\Gamma_o(q)$ are

formed from (3.3) by using the reasoning that leads to (5.1) which is valid for ferromagnetic coupling. (NB, in $\Gamma(q)$ the wave vector q is measured from the antiferromagnetic ordering wave vector, **w**.) After some algebra, one finds,

$$\Gamma(q) = a_{\circ}\kappa\,(1 + (q/\kappa)^2)(JT)^{1/2}\,, \tag{5.7}$$

and,

$$\Gamma_{\circ}(q) = 0.643\,(a_{\circ}\,q^2/\kappa)\,(JT)^{1/2}\,. \tag{5.8}$$

Hence, at the ordering wave vector the temperature dependence of the decay rate is provided by $\kappa\, T^{1/2}$, i.e. a slowing down occurs as the temperature tends to zero. Near the zone centre, for a fixed wave vector the decay rate increases with decreasing temperature.

6. Discussion

To the best of our knowledge, this is the first report of results from coupled-mode theory applied to the classical Heisenberg magnet in two dimensions. Given the unmatched success of coupled-mode theory to describe critical and paramagnetic fluctuations in three-dimensional magnets, there is, clearly, good reason to believe that the theory yields a reasonable account of fluctuations in the case of a two dimensional system. Looking at our results for the ferromagnetically and antiferromagnetically coupled systems, all features are in accord with physical intuition. However, as a caveat to our confidence in coupled-mode theory applied to a spatial dimension less than three recall that, the standard coupled-mode theory, which is used here, is known to fail in one dimension. In this case, it does not reproduce the weakly damped collective spin oscillations, reminiscent of linear spin waves, that are known to exist at low temperature. For our two-dimensional model, at the lowest temperature investigated, $\theta = 0.308$, there is no collective mode peak in $S(\mathbf{q},\omega)$ at $\mathbf{q} = \mathbf{w}\ (\mathbf{w}/2)$ for ferromagnetic (antiferromagnetic) coupling.

For a ferromagnetically coupled system the decay rate of long wavelength fluctuations is proportional to $q^2T^{1/2}$ if the wave vector, q, is large compared to the inverse correlation length, κ. In the other extreme, $q << \kappa$, there is a logarithmic correction to the expected q^2 - dependence, namely, $\Gamma(q) \alpha q^2 \{T \ln (\kappa/q)\}^{1/2}$. This implies that, for ferromagnetic coupling, the conventional theory of spin-diffusion does not apply. At the wave vector at which there is incipient antiferromagnet ordering, the decay rate decreases with decreasing temperature, and we find a temperature dependence given by $\kappa T^{1/2}$. Near the zone centre, the decay rate of the antiferromagnetically coupled system increases with decreasing temperature, namely, $\Gamma_o(q) \alpha q^2 T^{1/2} /\kappa$.

For a complete picture of the spin dynamics the coupled-mode theory must be analysed by a numerical method. We provide a comprehensive survey using a wide range of temperatures and wave vectors.

Acknowledgement

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Table 1

μ	θ	$a_0\kappa$
1.001	0.350	0.063
1.01	0.472	0.200
1.03	0.569	0.346
1.10	0.744	0.632
3.0	2.914	2.288

Representative values of the reduced temperature, $\theta = (3T/4J)$, and the inverse correlation length, κ, as a function of the dimensionless parameter μ that arises in the spherical model of spin correlations, cf. fig. (1).

Figure Captions

1. A graphical representation between the reduced temperature, $\theta = (3T/4J)$, and the parameter μ which arises in the spherical model; $1 = \theta I(\mu)$ where $I(\mu)$ is the extended Watson integral (3.7).

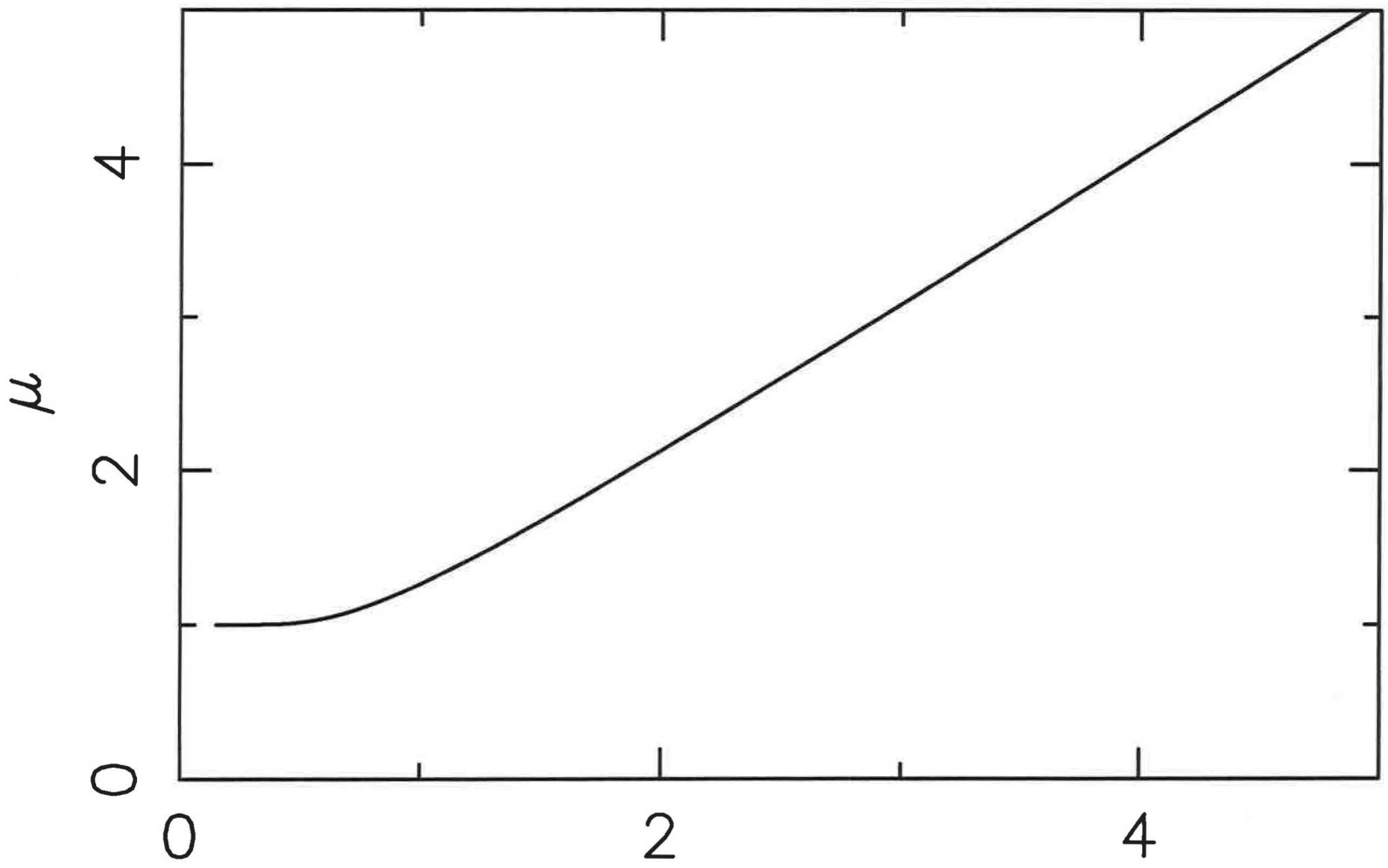
2. $F(\mathbf{q},t)$ is displayed as a function of time for three wave vectors, $\mathbf{q}_1$, $\mathbf{q}_2$, $\mathbf{q}_3$, specified in §4. Three temperatures are used, $\mu = 1.01$, 1.10 and ∞, cf. Table 1. The exchange coupling is antiferromagnetic. Solid line $\mu = \infty$; dashed line $\mu = 1.10$; dash-dot line $\mu = 1.01$. The energy constant satisfies $4J = 1$, and the unit increment of time in the plots is 1.330.

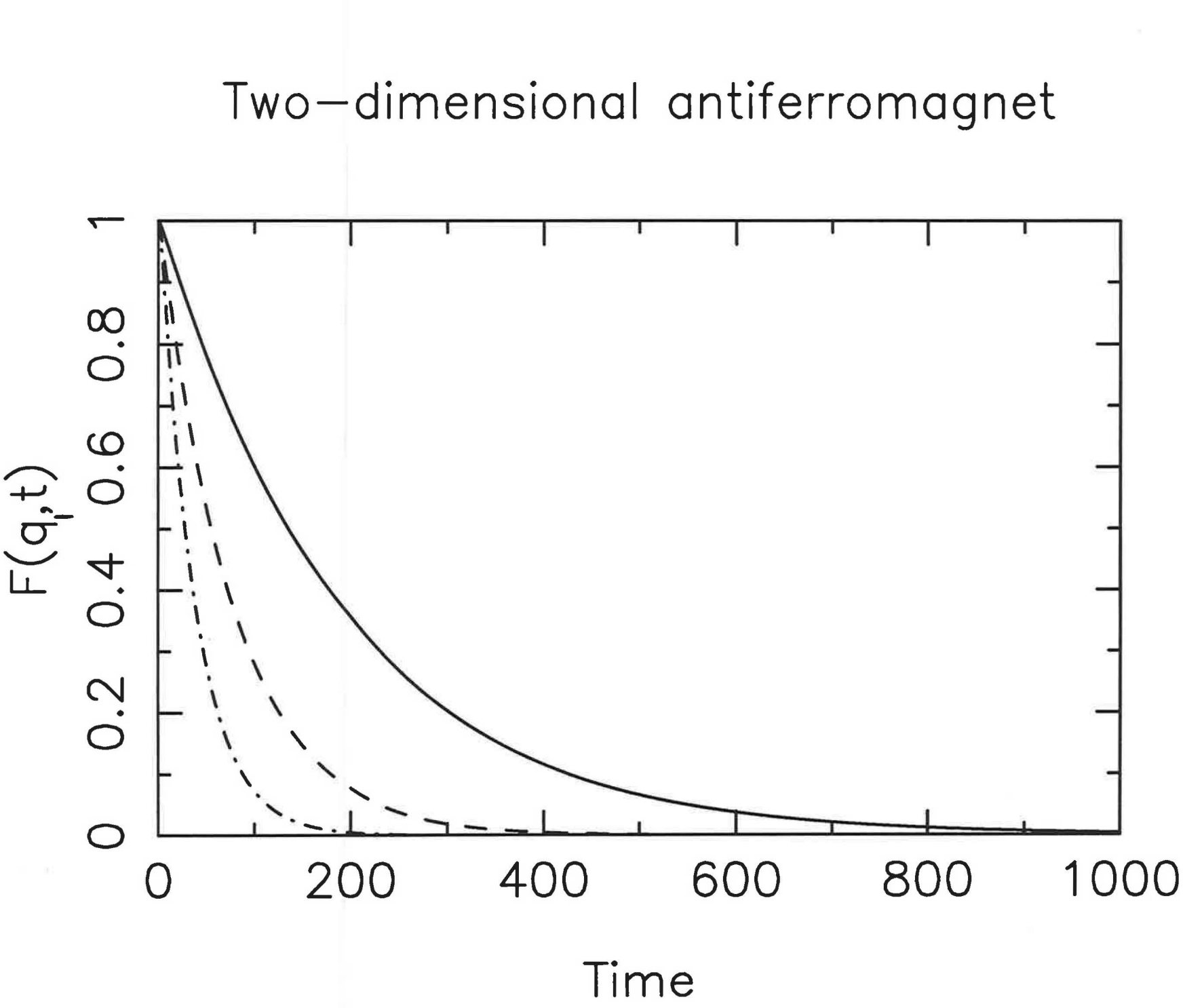
3. The response function (power spectrum) defined by (4.1) is displayed for the states used in fig. (2). The unit increment of frequency in the plots is 0.752, and $4J = 1$.

4. This figure and fig. (5) show $F(\mathbf{q},t)$ and $S(\mathbf{q},\omega)$ for a ferromagnetic exchange. The three wave vectors used are defined in §4. The temperatures are $\mu = 1.01(\theta = 0.472)$, $1.10(0.744)$, and $\mu = \infty$; the labelling of the different types of lines are defined in the caption to fig. (2).

5. Values of $S(\mathbf{q},\omega)$ obtained from the data for $F(\mathbf{q},t)$ shown in fig. (4).

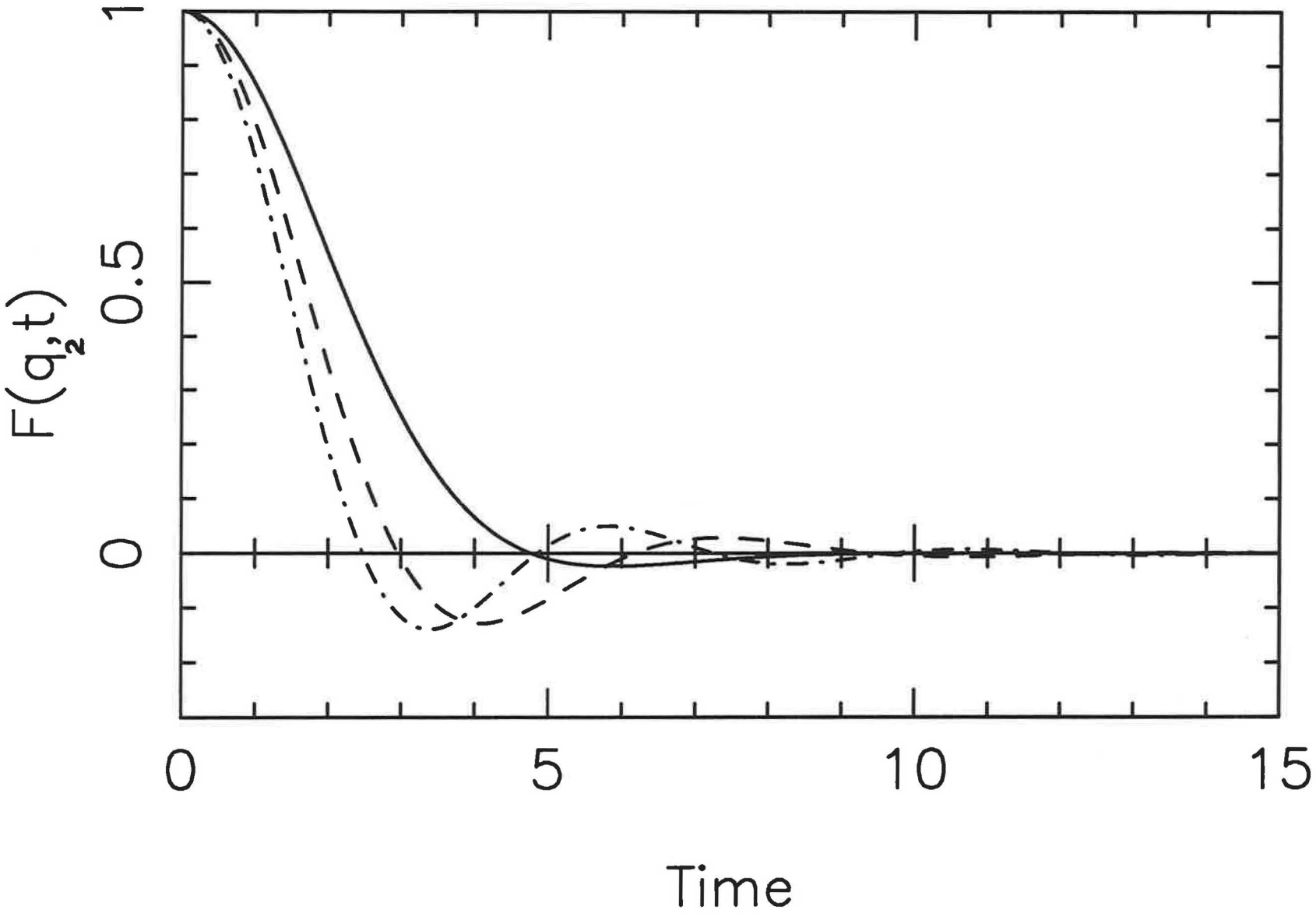
2−dimensional spherical model



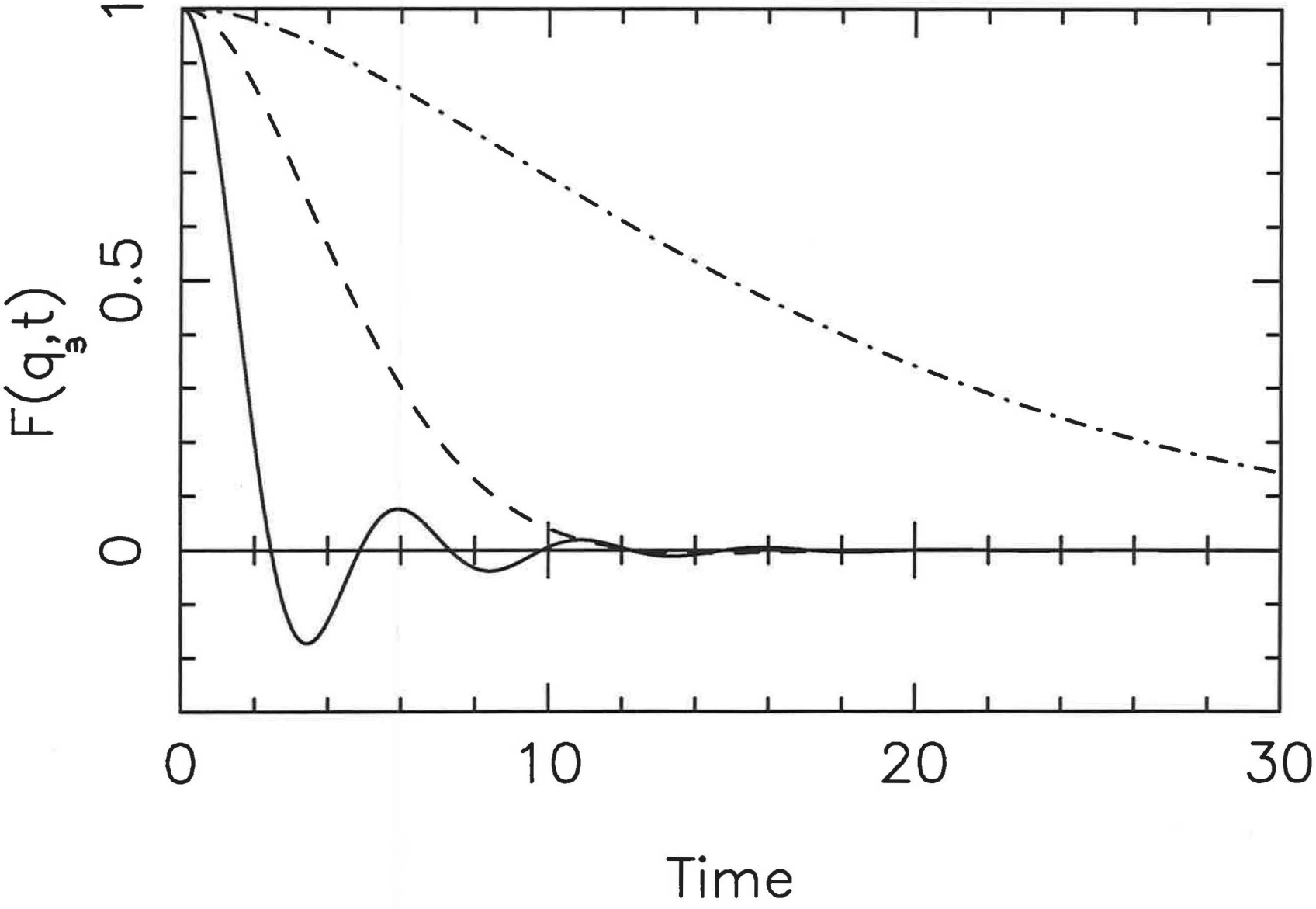

Two−dimensional antiferromagnet
F(q,t)
0
0.2
0.4
0.6
0.8
1
0
200
400
600
800
1000
Time

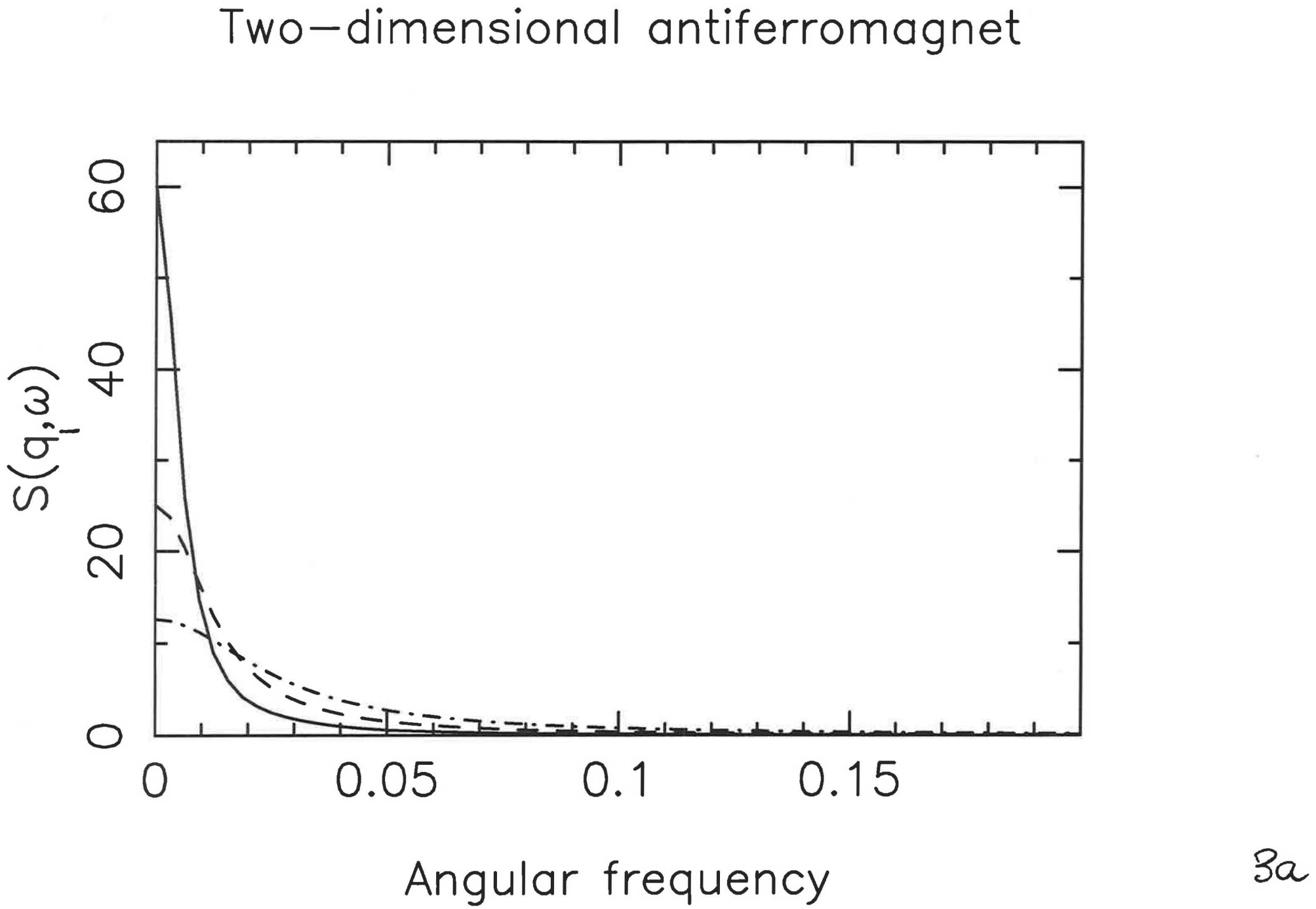
2a

Two–dimensional antiferromagnet

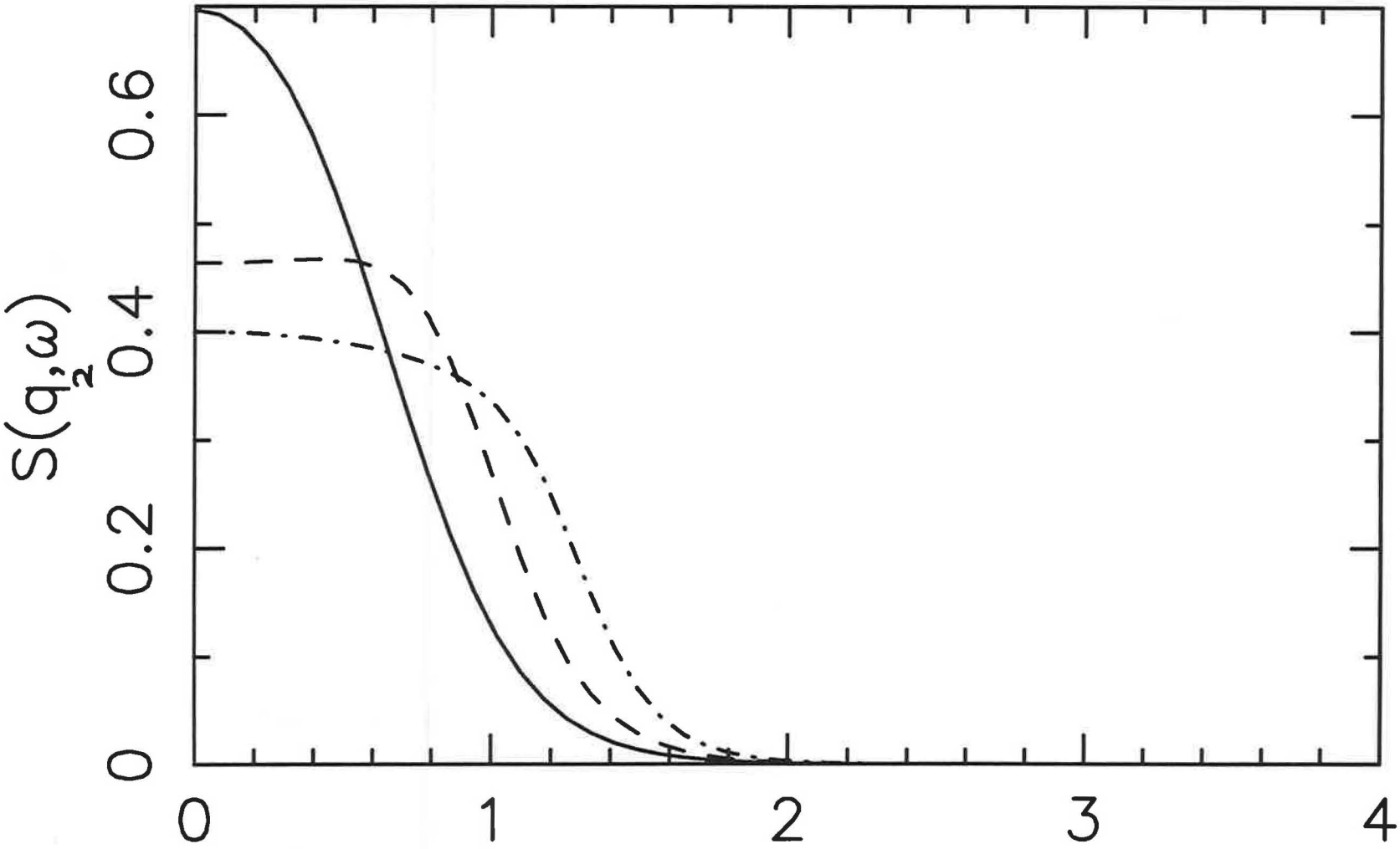


Two-dimensional antiferromagnet

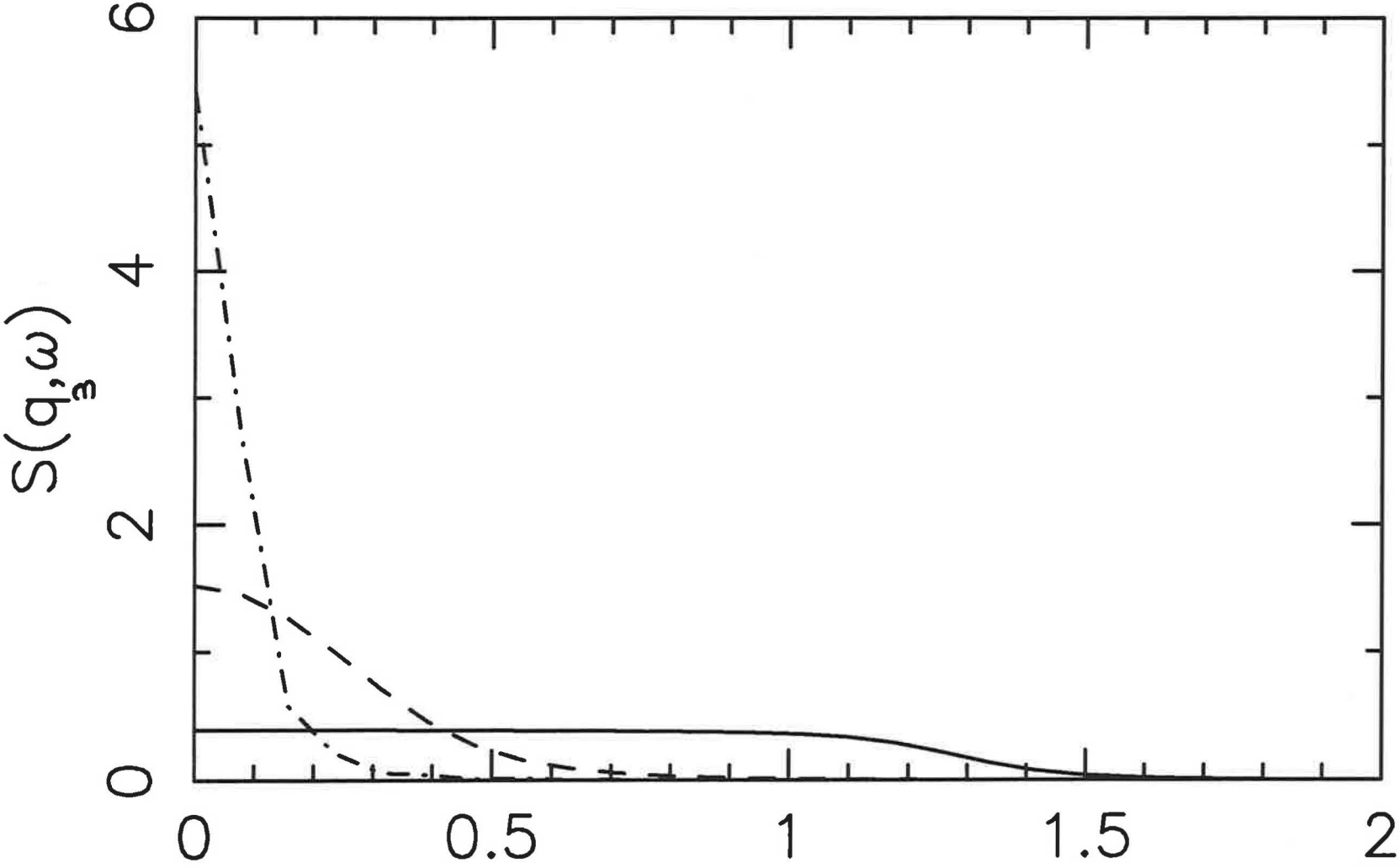



Two–dimensional antiferromagnet
S(q,ω)
0
20
40
60
0
0.05
0.1
0.15
Angular frequency

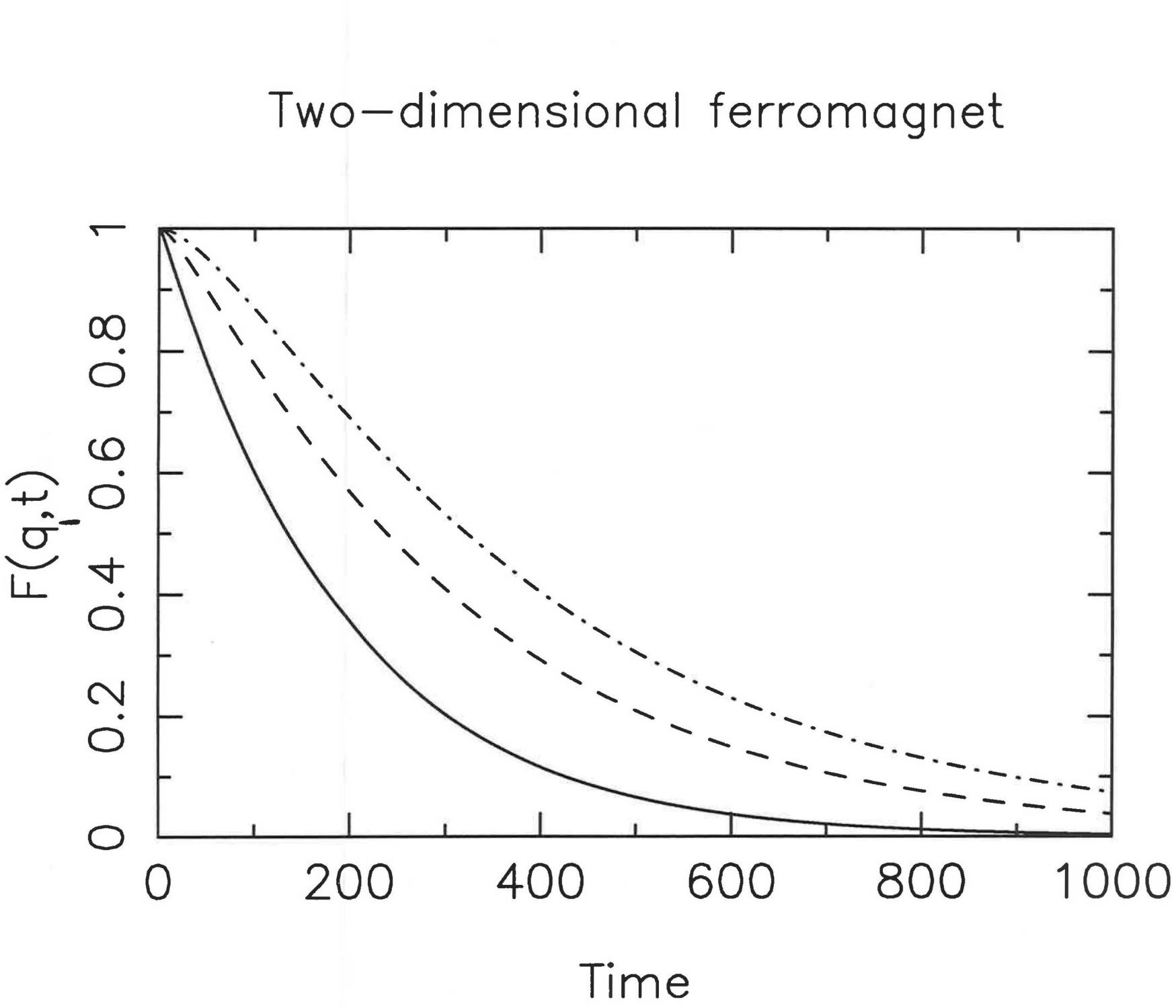
Two-dimensional antiferromagnet



Two-dimensional antiferromagnet

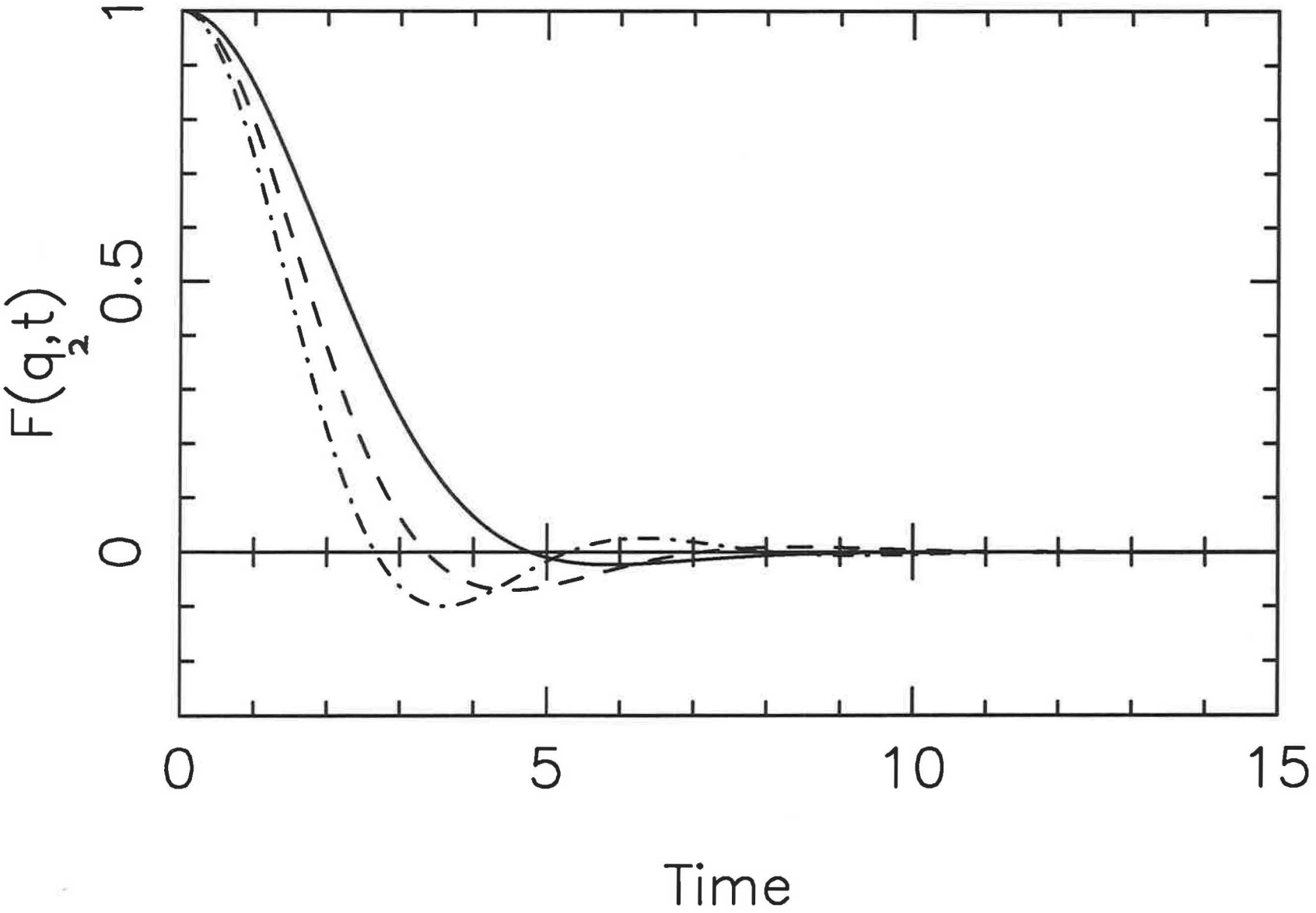


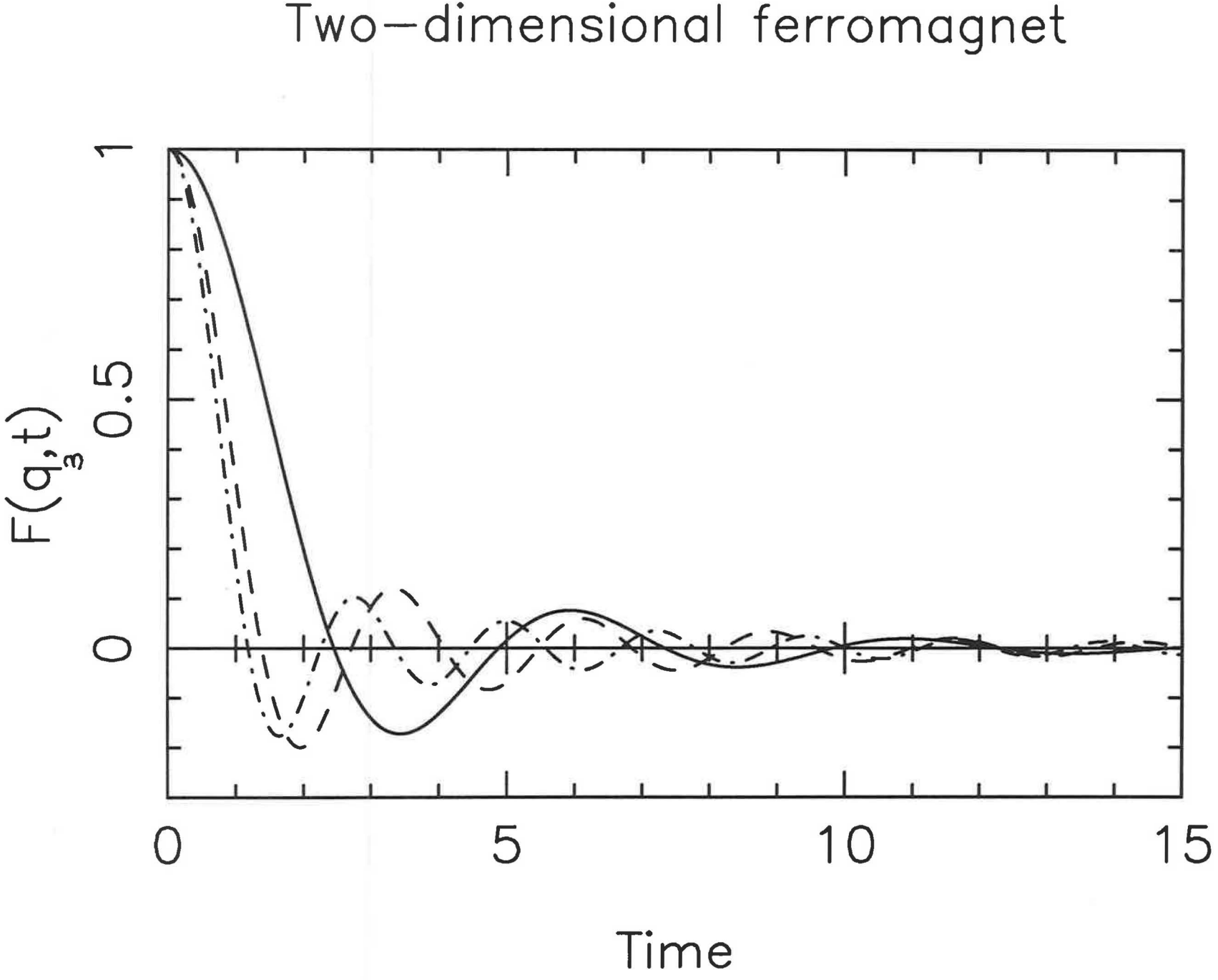
3c


Two−dimensional ferromagnet
F(q,t)
0
0.2
0.4
0.6
0.8
1
0
200
400
600
800
1000
Time

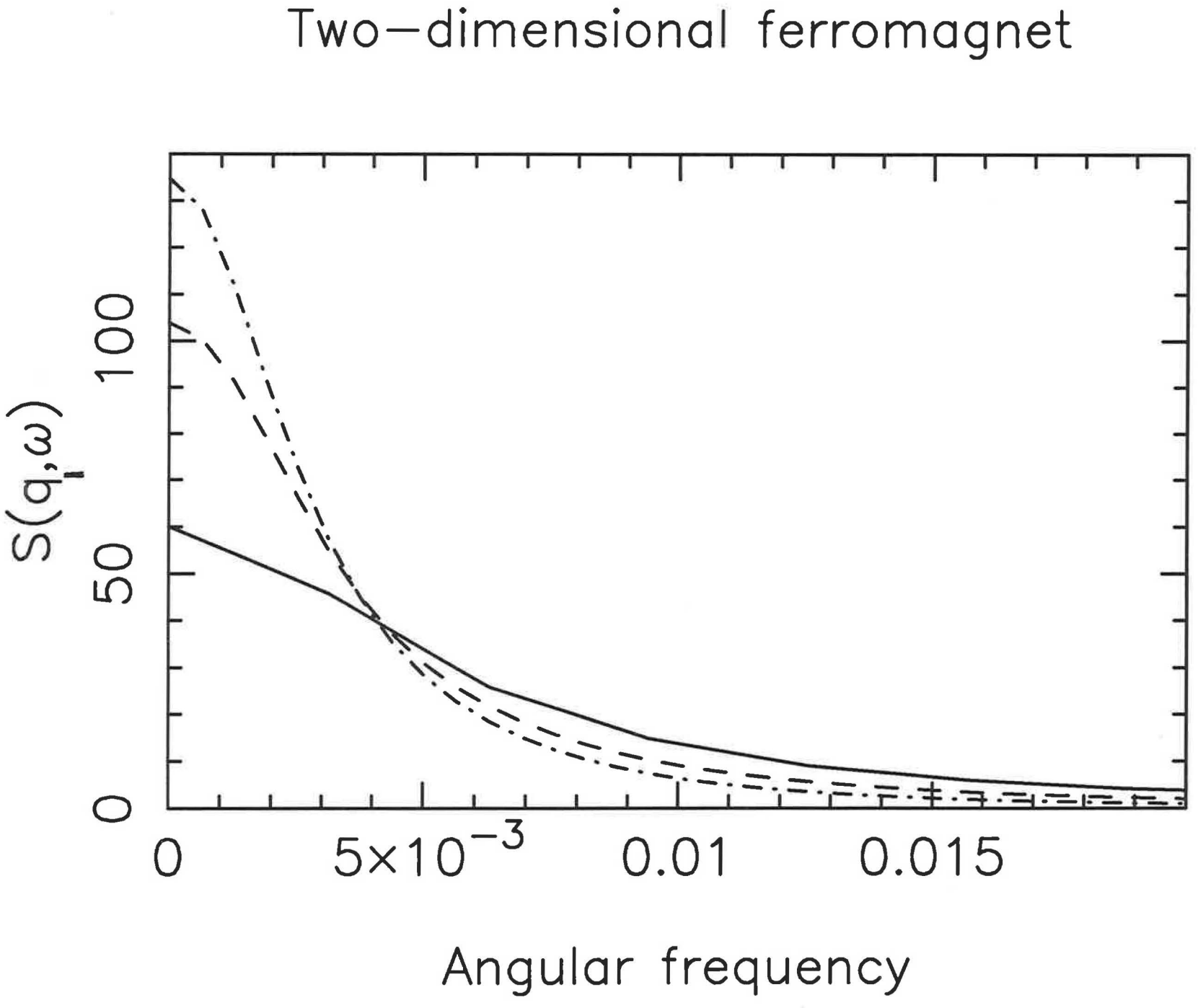
4a

Two–dimensional ferromagnet



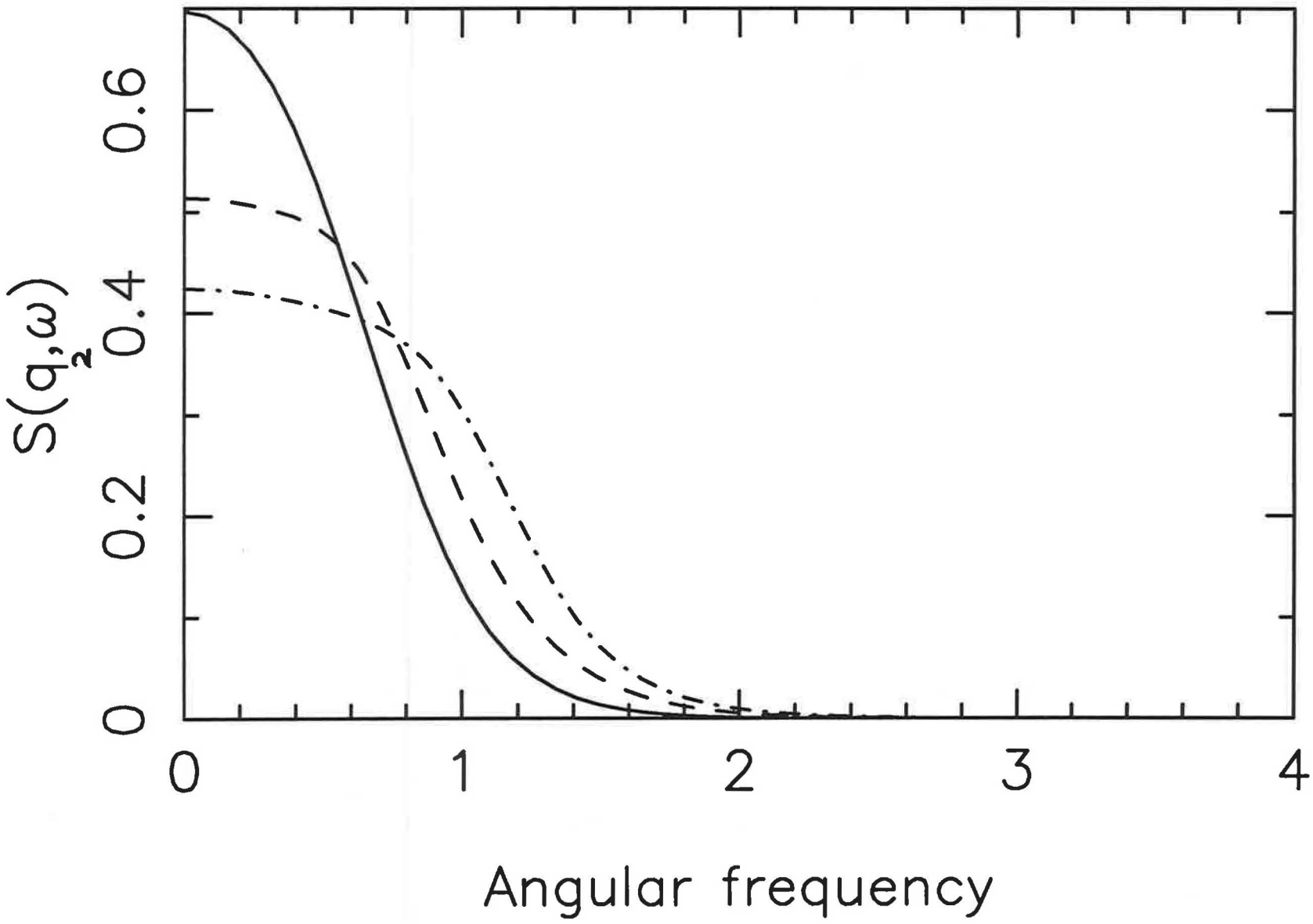


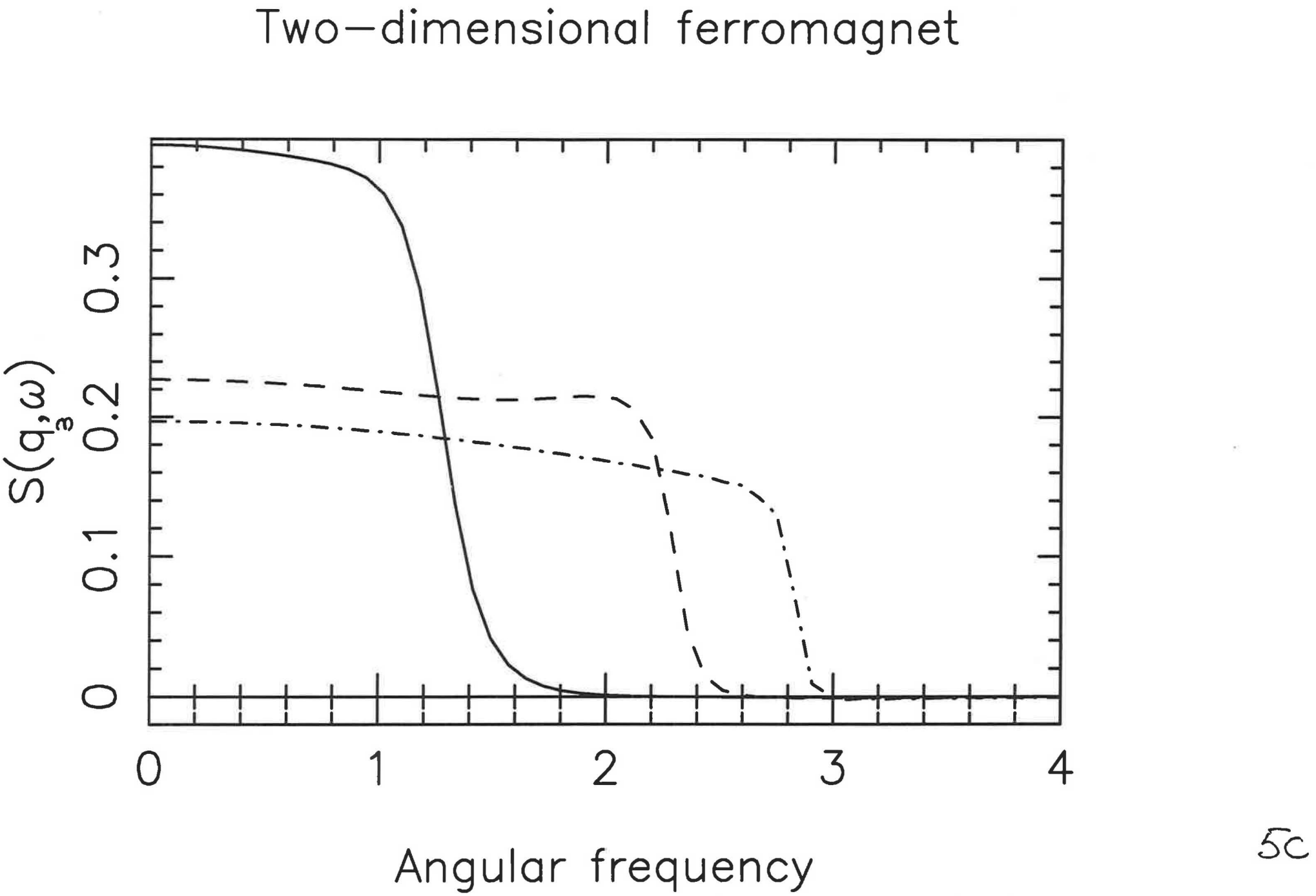
4c


Two-dimensional ferromagnet
S(q,ω)
0
50
100
0
5×10^-3
0.01
0.015
Angular frequency

5a

Two-dimensional ferromagnet




Two–dimensional ferromagnet
$S(q_3,\omega)$
0
0.1
0.2
0.3
0
1
2
3
4
Angular frequency

5c