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Time-Dependent Spin Correlations in the Heisenberg Magnet at Infinite Temperature

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Abstract

A coupled-mode theory of spin fluctuations in the d-dimensional Heisenberg magnet at infinite temperature is used to predict the time-dependence of various spin correlation functions. The real-space spin autocorrelation function is shown to have a long-time behaviour ~ $(1/t)^{d/\theta}$ where $\theta = (4 + d)/2$. Properties at intermediate values of the time are extracted from the theory by numerical analysis. In this time window, the reciprocal-lattice spin autocorrelation function, $G(q,t)$, is, to a good approximation, an exponential function of time. The decay rate is proportional to q^{α}, where q is the wave vector. Analysis of our numerical data indicates that, the exponent α depends weakly on d, and it is significantly different from the value 2 which is compatible with a spin diffusion model. In the asymptotic limit, defined by $q \to 0$, $t \to \infty$ and $q^2 t \to 0$, $G\,(q,t)$ is a function of a single variable = (tq^{θ}). This result rules against the validity of a diffusion model also in the asymptotic limit.

We report in this Letter calculations of time-dependent spin correlations in the classical Heisenberg magnet at infinite temperature. In the model magnet, unit vector spins, $\{\mathbf{S}_a\}$, are arranged on a lattice of dimension d, where d = 1,2 or 3. The quantity used to formulate the calculations is the spin autocorrelation function,

$$G(\mathbf{q},t) = \langle \mathbf{S}(\mathbf{q},t) \cdot \mathbf{S}(-\mathbf{q},0) \rangle, \tag{1}$$

in which $\mathbf{S}(\mathbf{q})$ is a spatial Fourier component of the spin density, t is the time variable, and $G(\mathbf{q},0) = 1$. Values for $G(\mathbf{q},t)$ are obtained from the so-called coupled-mode theory of spin fluctuations. It is shown here that, according to this theory, $G(\mathbf{q},t)$ at both intermediate and asymptotic ($t \to \infty$) times is not consistent with the conventional spin diffusion model. Instead, for asymptotic times we predict a power law decrease of the spin-spin correlation function,

$$\langle \mathbf{S}_a(t) \cdot \mathbf{S}_a(0) \rangle \sim (1/t)^{d/\theta}, \tag{2}$$

where $\theta = (4 + d)/2$, while at intermediate times,

$$\langle \mathbf{S}_a(t) \cdot \mathbf{S}_a(0) \rangle \sim (1/t)^{d/\alpha}, \tag{3}$$

and numerical analysis suggests that α depends weakly on d and takes values $\sim 1.62 - 1.90$, cf. Table 1. By contrast, the conventional spin diffusion model leads to a power law decrease with an exponent = $(d/2)$.

Studies of time-dependent spin correlations in the isotropic three-dimensional Heisenberg magnet at high temperatures appear to date back to Van Vleck's work in 1939. He drew attention to the potential value of neutron scattering experiments on paramagnets as a means of determining the magnitude of the exchange interaction in the sample. Later work by De Gennes (1958) marked the beginning of our appreciation of the subtle wave vector dependence of the neutron cross-section, which is proportional to the time Fourier transform of $G(\mathbf{q},t)$, i.e. the power spectrum. The findings from this and related work, similarly based on a study of frequency moments of the power spectrum, are reviewed by Marshall and Lowde (1968). Thereafter came seminal work by Wegner, and others, that provides a detailed theory of critical and paramagnetic spin fluctuations; copious references are given in a review by Kawasaki (1975).

A resurgence of interest in the time-dependent properties of the Heisenberg magnet at infinite temperature is underway, partly because of intriguing results from computer simulations on one-dimensional (1D) systems, e.g. Srivastava et al. (1994). Findings from such studies do not support the standard spin diffusion phenomenology. Applied to the autocorrelation function, $G(q,t)$, a spin diffusion mechanism leads to,

$$G(q,t) \sim \exp(-Dq^2 t) \, ,$$

for a sufficiently small wave vector, q, and a sufficiently long time t. At the moment, the authors of the reports of computer simulation studies do not have a consensus view as to the correct form for $G(q,t)$. In part, at least, this stems from the fact that these results depend on the time window sampled in the data analysis.

An alternative line of investigation is to apply at infinite temperature the theory developed by Wegner for 3D systems. Following this line, Lovesey and Balcar (1994) have shown that coupled-mode theory, as it is now normally called, applied to a 1D system predicts different, although almost deceivingly similar, properties at intermediate and asymptotic times. Their results for the spin-spin correlation function, given by (2) and (3) evaluated for $d = 1$, are indeed strikingly similar to results from some computer simulation studies (Gerling and Landau 1989, 1990).

Returning, for the moment, to 3D systems Chertkov and Kolokolov (1994) predict the long-time behaviour of the spin-spin correlation function using a new theory of spin fluctuations, which seems to be different from coupled-mode theory. Citing work by Blume and Hubbard (1970), they claim that the exponent for (2) deduced from coupled-mode theory is (3/2), while they obtain from the new theory a value (6/7). In fact, this latter exponent is the same as the expression (d/θ), appearing in (2), evaluated for $d = 3$. Previous work with 3D coupled-mode theory at infinite temperature is incomplete because it assumes the existence of a conventional spin diffusion mechanism.

The Heisenberg model is defined by the Hamiltonian,

$$\mathcal{H} = \tfrac{1}{2} J \sum_{a,b,}^{n.n.} \mathbf{S}_a \cdot \mathbf{S}_b \,, \tag{4}$$

in which nearest-neighbour spins are coupled by an exchange interaction of strength J. The spin autocorrelation function, $G(\mathbf{q},t)$, is obtained from a memory function, $K(\mathbf{q},t)$, which is derived from (4) by a standard method of approximation which constitutes the coupled-mode theory previously referred to. One finds (see, for example, Lovesey 1986),

$$K(\mathbf{q},t) = \tfrac{1}{3}(rJ)^2 \, \tfrac{1}{N} \sum_{\mathbf{k}} \, (\gamma_{\mathbf{k}} - \gamma_{\mathbf{q-k}})^2 \; G(\mathbf{k},t) G(\mathbf{q}-\mathbf{k},t), \tag{5}$$

while $G(\mathbf{q},t)$ satisfies,

$$\partial_t \; G(\mathbf{q},t) = -\int_0^t \, \mathrm{d}t' \; G\,(\mathbf{q},t') K(\mathbf{q},\, t-t'). \tag{6}$$

In (5), r is the number of nearest neighbours on the lattice, e.g. $r = 4$ for $d = 2$, and $\gamma_{\mathbf{q}}$ is a geometric factor generated from the point-group symmetry of the lattice, and for a square lattice ($d = 2$),

$$\gamma_{\mathbf{q}} = (1/2)(\cos a_o q_x + \cos a_o q_y),$$

where a_o is the lattice spacing. Equations (5) and (6) for $d = 1$ are identical with those analysed by Lovesey and Balcar (1994) when their renormalization parameter, μ_q, is set equal to unity for all values of the wave vector.

Asymptotic properties ($q \to 0$, $t \to \infty$, $q^2 t \to 0$) of the autocorrelation and memory functions are obtained from consideration of homogeneous forms,

$$G(q,t) = \mathrm{G}\,(q\,\lambda^a, t\,\lambda^b),$$

and (7)

$$K(q,t) = \lambda\, K\,(q\lambda^a, t\lambda^b),$$

taken in the limit $\lambda^a \to 0$. From (6) one finds the exponent $b = 1/2$. The second exponent, a, depends on dimensionality, and it is derived from (5). In the asymptotic limit, the latter reduces to the form,

$$K(q,t) \sim q^2 \int_q k^{d+1}\, \mathrm{d}k\, G^2(k,t)\,, \tag{8}$$

and from this expression and (7), $a = -1/(4+d)$. Hence, in the asymptotic limit,

$$G(q,t) = g(tq^\theta)\,,$$

and, (9)

$$K(q,t) = q^{2\theta} f(tq^\theta)\,,$$

where the exponent $\theta = (4 + d)/2$, and the scaling functions $f(x)$ and $g(x)$ are determined by coupled integral equations derived from (6) and (8).

However, several interesting results follow directly from (8) and (9). For example, in the asymptotic limit,

$$\acute{K}(q,t) \sim q^2 \, (1/t)^{(d+2)/\theta} \, , \tag{10}$$

and $G(q,t)$ shares the same power law decay with time. The result (2) for the spin-spin correlation function follows from the homogeneous form of the autocorrelation function and,

$$< \mathbf{S}_a(t) \cdot \mathbf{S}_a(0) > \sim \int q^{d-1} \, \mathrm{d}q \, G(q,t) \, .$$

The exponent of $(1/t)$ in the spin-spin correlation function, d/θ evaluated for $d = 1$ and $d = 3$ agrees with the results reported by Lovesey and Balcar (1994) and Chertkov and Kolokolov (1994), respectively. It is interesting to observe that, if the Kubo formula is valid (10) implies a finite diffusion constant, i.e.,

$$D = (1/q^2) \int_0^{\infty} \mathrm{d}t \, \exp(-st) \, K(q,t) \, ,$$

evaluated with the limits $q \to 0$, followed by $s \to 0$.

Properties of $G(q,t)$ and $K(q,t)$ at intermediate times, $5 \leq t\,J \leq 100$, and small wave vectors have been deduced from a numerical analysis of (5) and (6). Figs. (1) and (2) show for $d = 2$, $\log G$ vs. t, and $\log K$ vs. $\log t$. It is evident from the data that, to a very good approximation, $G(q,t)$ is an exponential function of time, and $K(q,t)$ has an inverse power law dependence on time. Similar behaviour for $d = 1$ has been reported by Lovesey and Balcar (1994), and new calculations for $d = 3$ also display the same functional forms.

Hence, for $d = 1,2$ and 3 numerical data for the spin autocorrelation function evaluated for a small wave vector and intermediate times are compatible with,

$$G(q,t) \sim \exp(-\Gamma t q^{\alpha}) . \tag{11}$$

Given this expression for $G(q,t)$ one arrives at the result (3) for the spin-spin correlation function. The result for $K(q,t)$ which corresponds to (11) for $G(q,t)$ is convenient for obtaining an estimate of the exponent α. Using (11) in (8),

$$K(q,t) \sim (1/t)^{(d+2)/\alpha} , \tag{12}$$

and, for d = 2, fig. (2) shows that this can be accurately fitted to the numerical data. Values of α obtained by fitting (12) to the data for d = 1, 2 and 3 are gathered in Table 1. Comparing results for the spin-spin correlation function at intermediate and asymptotic times we conclude that the exponent of $(1/t)$ decreases as one moves the time window to large values of t, and the effect is most pronounced in the largest spatial dimension.

Note that the refine coupled-mode theory for d = 1 used by Lovesey and Balcar (1994) when compared to the standard version of the theory used here leads to exactly the same asymptotic results, and different results at intermediate times. For example, the refined theory (d = 1) gives α = 1.50, cf. Table 1 based on the standard theory.

To round off our discussion of the Heisenberg model at infinite temperature, we consider the behaviour of the autocorrelation function for large wave vectors, near the Brillouin zone boundary. For d = 1, Lovesey and Balcar (1994) report oscillations in $G(\pi/a_0,t)$ as a function of time. The corresponding power spectrum, $S(\mathbf{q},\omega)$, which is proportional to the cross-section for inelastically scattered neutrons, is consequently significantly different from a gaussian function of ω. Figs. (3) and (4) show $G(\mathbf{q},t)$ and $S(\mathbf{q},\omega)$ for d = 2 and 3, and $\mathbf{q}$ at the zone boundary. It is evident that, for both lattices, $G(\mathbf{q},t)$ is strongly oscillatory as a function of time. The departure of $S(\mathbf{q},\omega)$ from a gaussian function of ω is most pronounced for d = 3. This might be expected since the mean square width of $S(\mathbf{q},\omega)$ increases with d. The second frequency moment is,

$$\omega_o^2(\mathbf{q}) = K(\mathbf{q},0) = \tfrac{2}{3}\, rJ^2(1-\gamma_{\mathbf{q}}),$$

and $\gamma_{\mathbf{q}} = -1$ at the Brillouin zone boundary.

Acknowledgement

One of us (S.W.L.) wishes to thank Prof. H. Lesche for correspondence, and the observation, verified by the present work, that the exponent for the spin-spin function obtained by Chertkov and Kolokolov (1994) for $d = 3$ is compatible with the exponent for $d = 1$ derived by Lovesey and Balcar (1994). A.C. and V.T. are grateful for the support provided by the Rutherford Appleton Laboratory in the period during which the work was completed.

Numerical Analysis of $K(q,t)$ at intermediate times: $K(q,t) \sim (1/t)^{(d+2)/\alpha}$

d	$(a_o q/\pi)$	α
1	1/20	1.62
2	1/24	1.88
3	1/12	1.89

Table 1

Values of the quantity α deduced from numerical results for $K(q,t)$ at intermediate values of the time t, and a small value of q which is listed in the table. Results are given for the three lattices considered in the text: $d = 1$ (linear chain), $d = 2$ (square lattice), and $d = 3$ (simple cubic lattice). The quantity a_o is the lattice spacing. See also fig. (2). In the asymptotic limit, the quantity α is replaced by $\theta = (4 + d)/2$.

References

- Blume, M., and Hubbard, J., Phys. Rev. **B1** (1970) 3815

- Chertkov, M., and Kolokolov, I., Phys. Rev. **B49** (1994) 3592

- De Gennes, P. G., Phys. and Chem. Solids **4** (1958) 223

- Gerling, R.W., and Landau, D.P., Phys. Rev. Lett. **63** (1989) 812

- " " Phys. Rev. **B42** (1990) 824

- Kawasaki, K., in Phase Transitions and Critical Phenomena, vol. 5a, edited by C. Domb and M.S. Green (Academic Press, London, 1975)

- Lovesey, S.W., and Balcar, E., J. Phys.:Condens. Matter **6** (1994) 1253

- Lovesey, S.W., Condensed Matter Physics: Dynamic Correlations, Frontiers in Physics vol. 61 (Benjamin/Cummings Publishing Co., Menlo Park, 1986)

- Marshall, W., and Lowde, R.D., Rept. Prog. Phys. **B31** (1968) 705

- Srivastava, N., Liu, J. - M., Viswanath, V.S., and Müller, G., J. Appl. Phys. to appear

- Van Vleck, J.H., Phys. Rev. **55** (1939) 924

Figure Captions

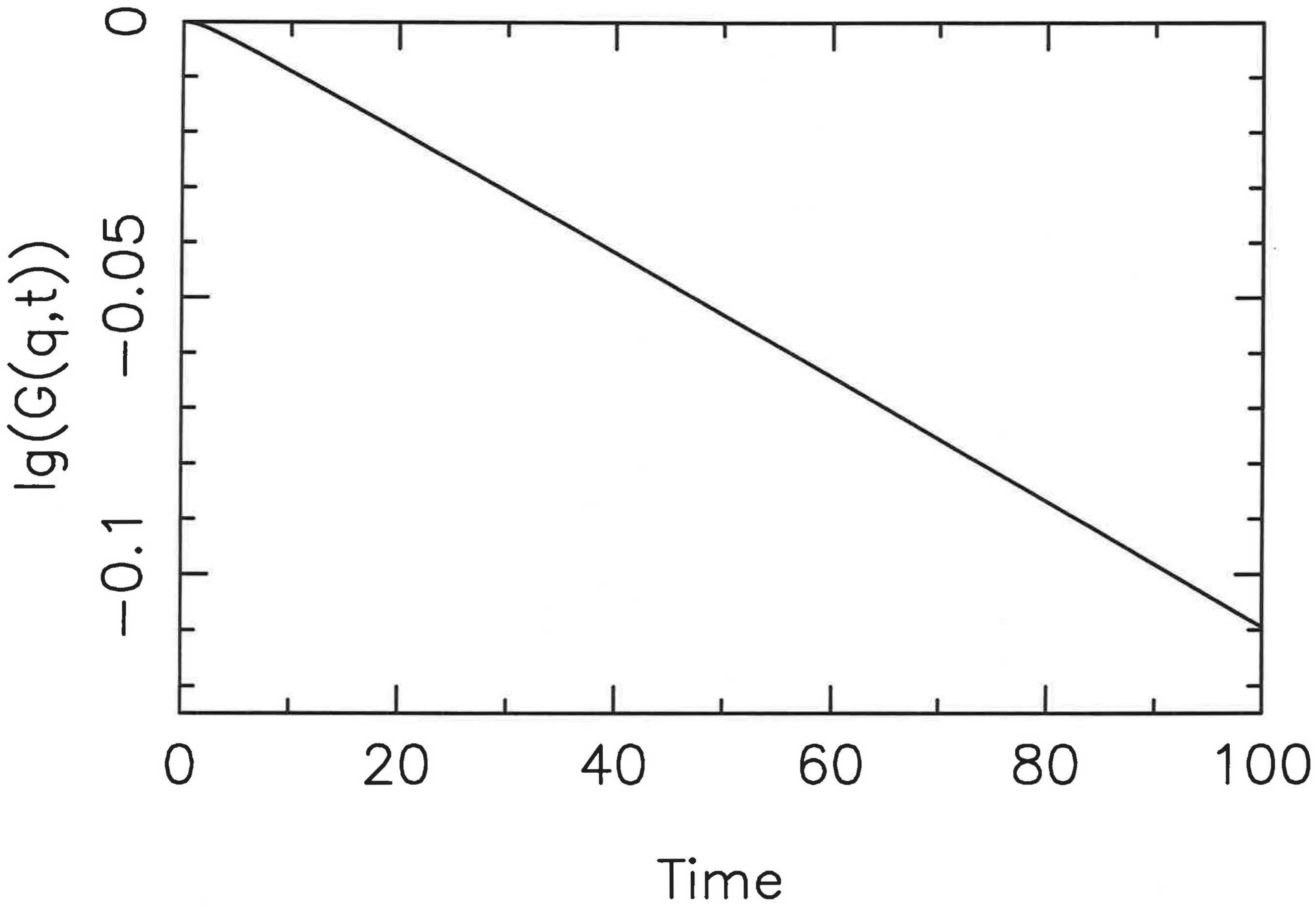
(1) The spin autocorrelation function, $G(q,t)$ derived from (5) and (6) is displayed in the form log G vs. t. Results are for a square (d = 2) lattice and a wave vector $q = (\pi/24a_o)$; $\mathrm{q} = (\pi/a_o)$ (0,1/24). The unit of energy ($\hbar = k_B = 1$) is $4J = 1$, and the unit increment of time = 1.330.

(2) The memory function, $K(q,t)$, which corresponds to the data in fig. (1) for $G(q,t)$ is displayed in the form log K vs. log t. The slope of the linear portion of the curve, which extends from log t = 0.3 to log t = 2.0, is found to be 2.13. Hence, from (12) one finds α = (4/ 2.13) = 1.88.

(3) The spin autocorrelation function, $G(\mathbf{q},t)$, is displayed as a function of time for d = 2 (square lattice) and the wave vector at the zone boundary, $\mathbf{q} = (\pi/a_o)(1,1)$. The accompanying figure is the power spectrum, $S(\mathbf{q},\omega)$, which is proportional to the cross-section for inelastic neutron scattering. Units are specified in the caption to fig. (1).

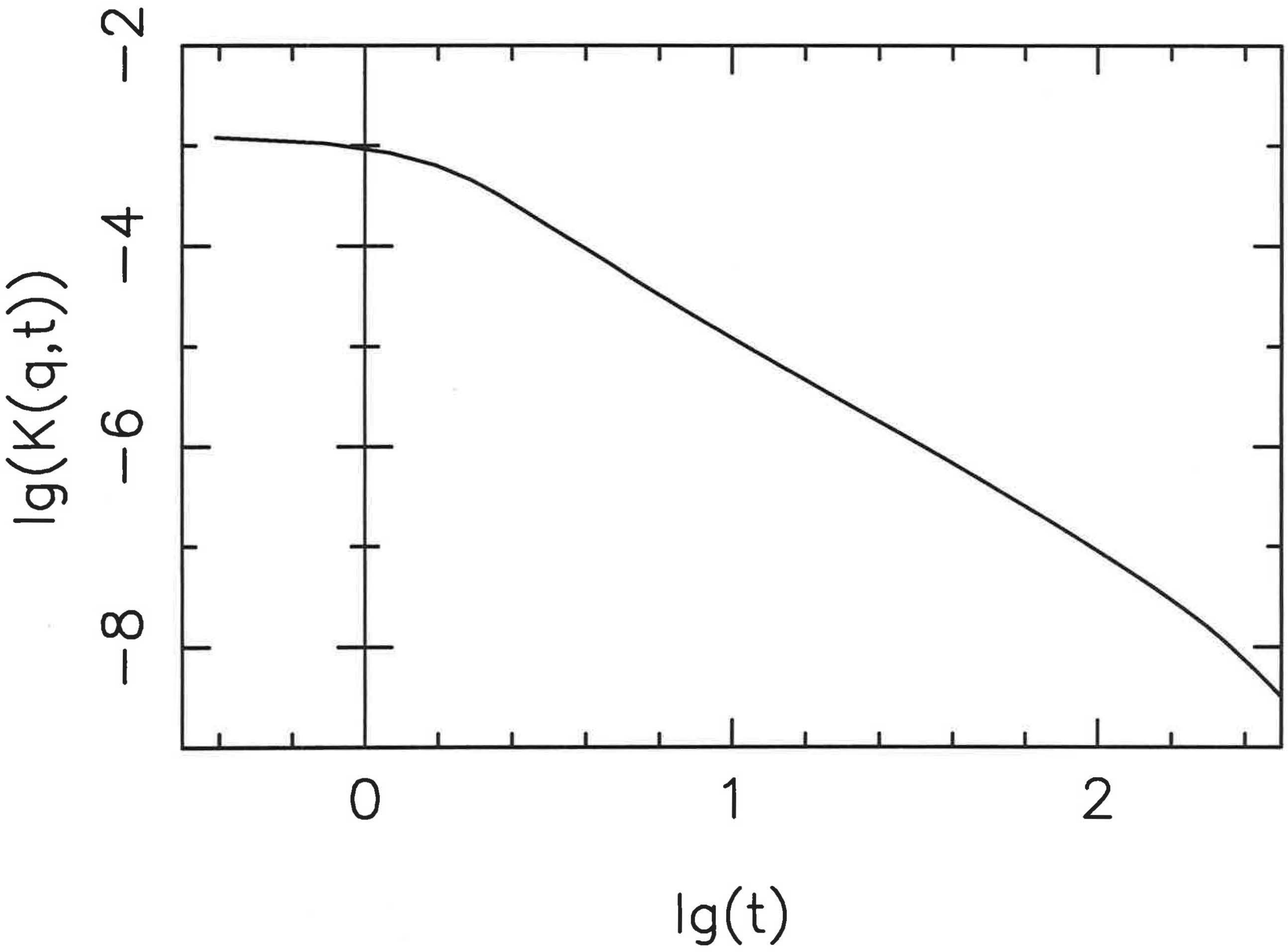
(4) The quantities shown in fig. (3) are given for d = 3 (simple cubic lattice) and $\mathbf{q} = (\pi/a_o)(1,1,1)$. The unit of energy is $6J$ = 1, and the unit increment of time = 1.637.

Two-dimensional s.s. magnet



1

Two-dimensional s.s. magnet



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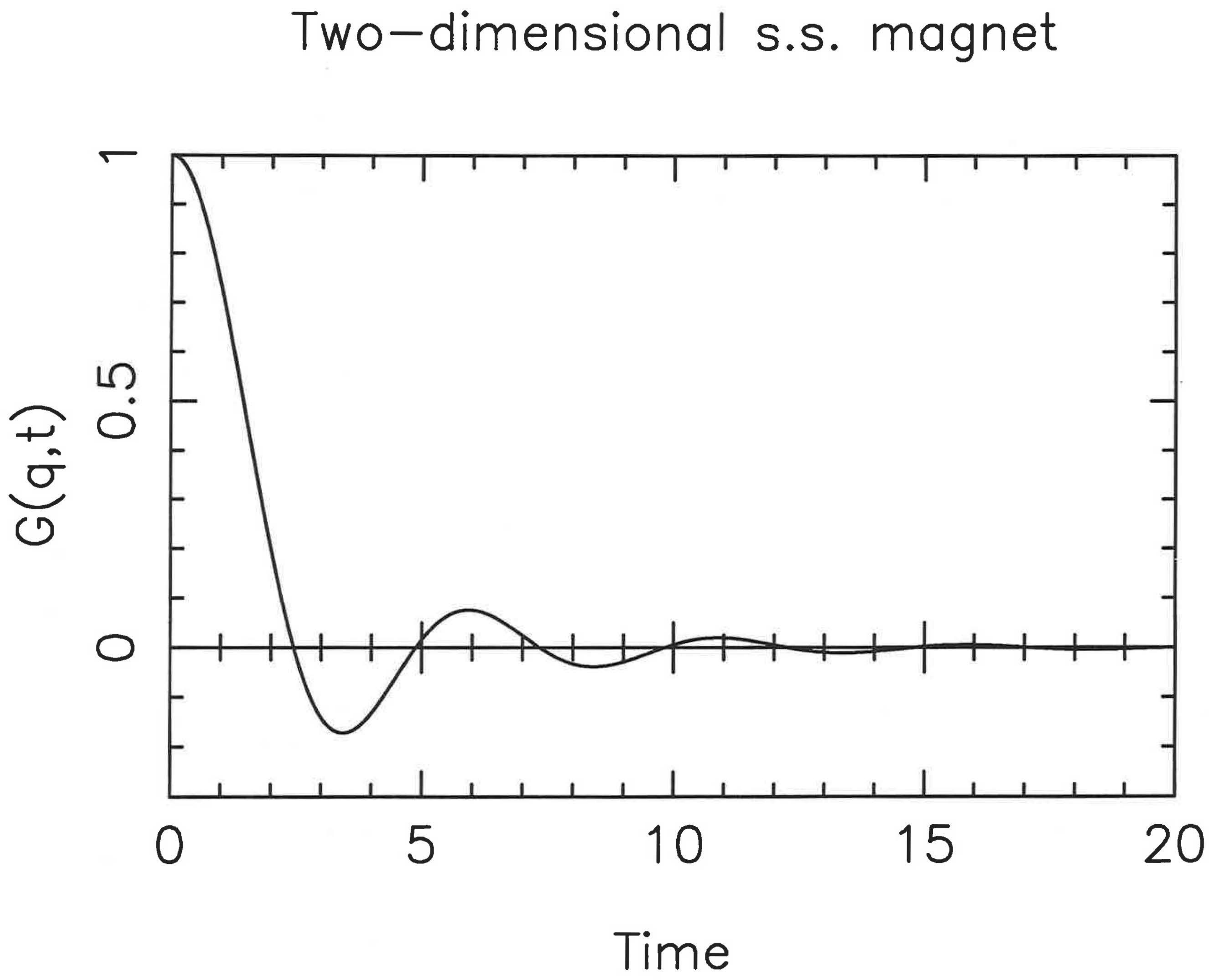
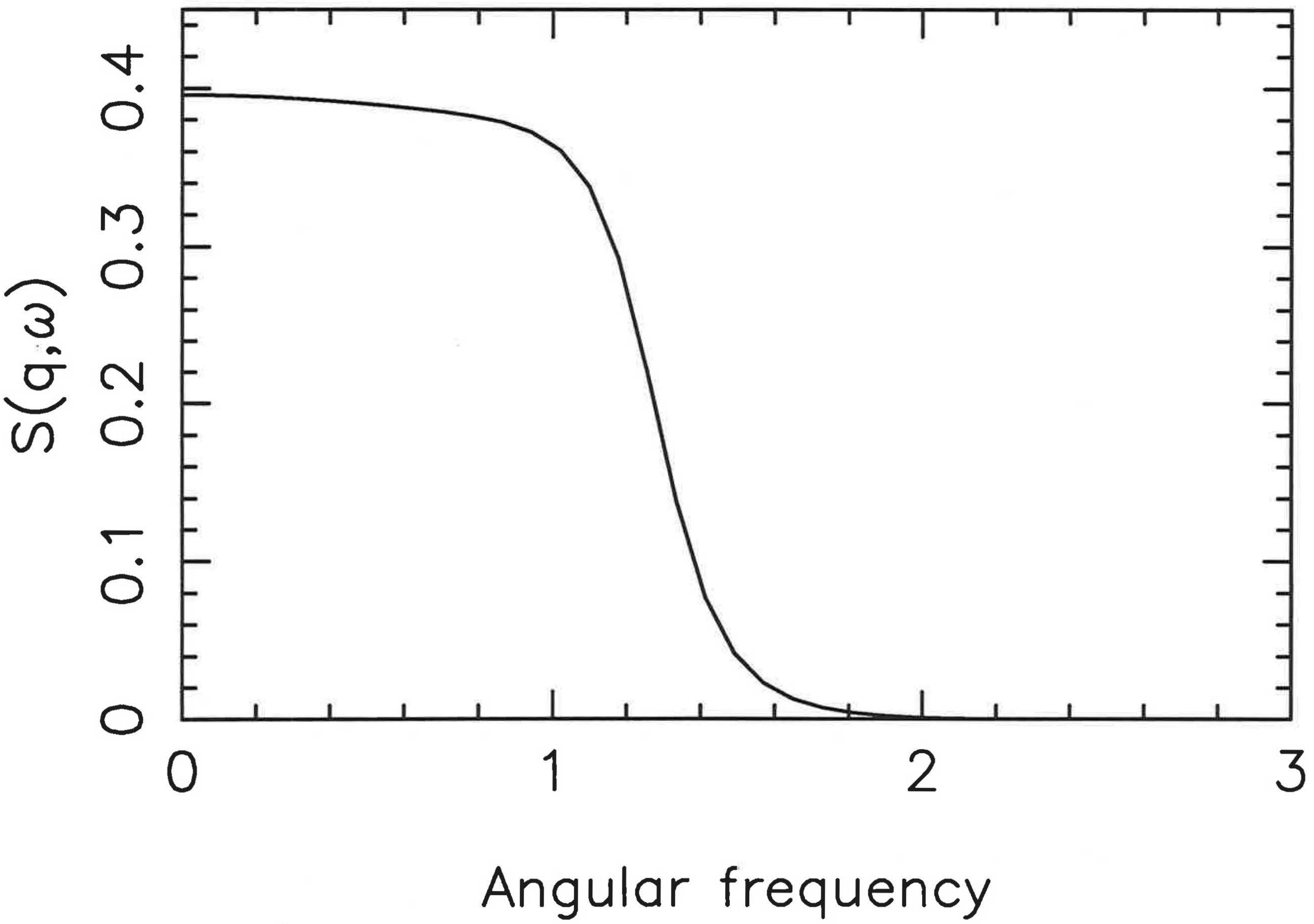


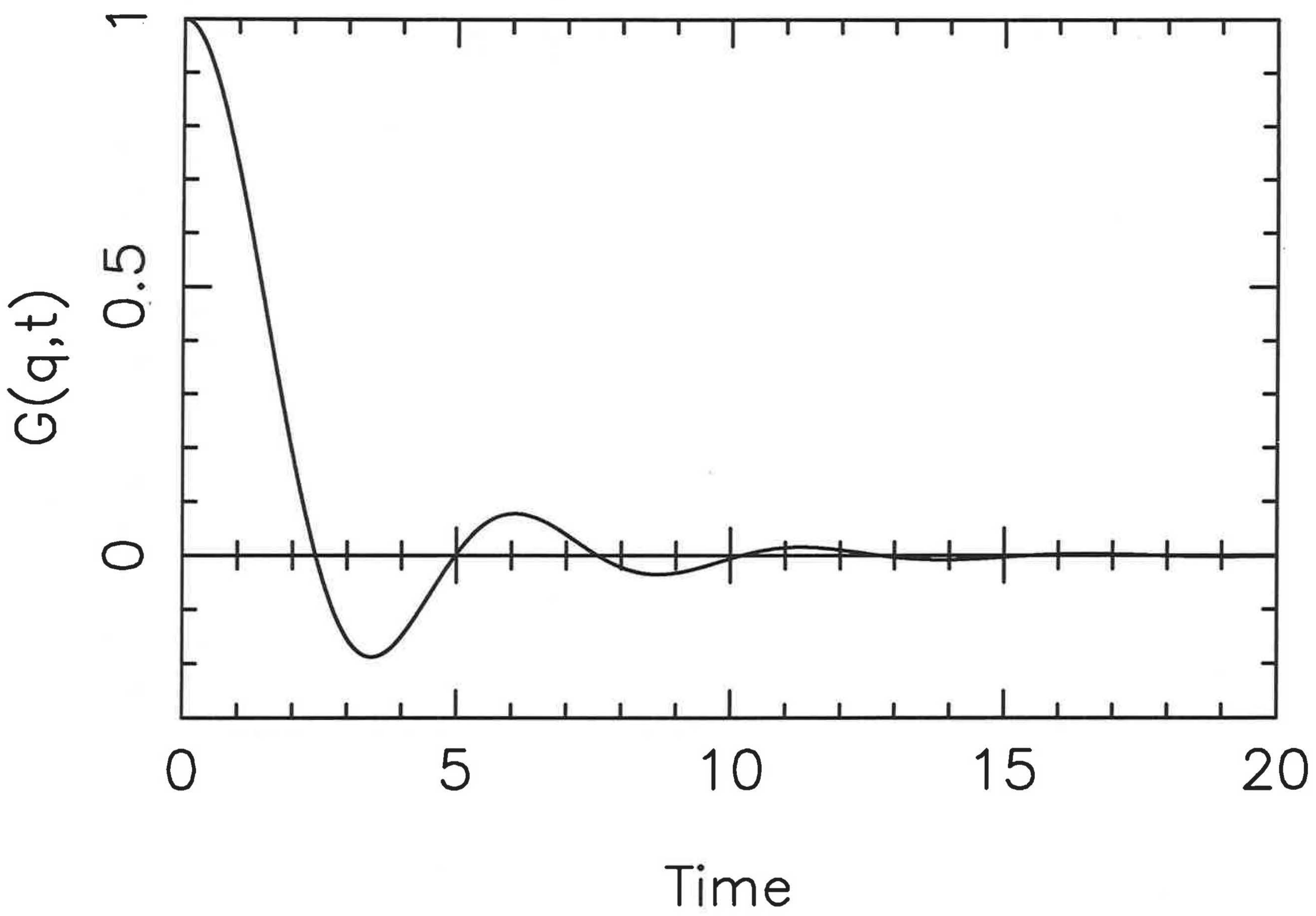
3a

Two-dimensional s.s. magnet

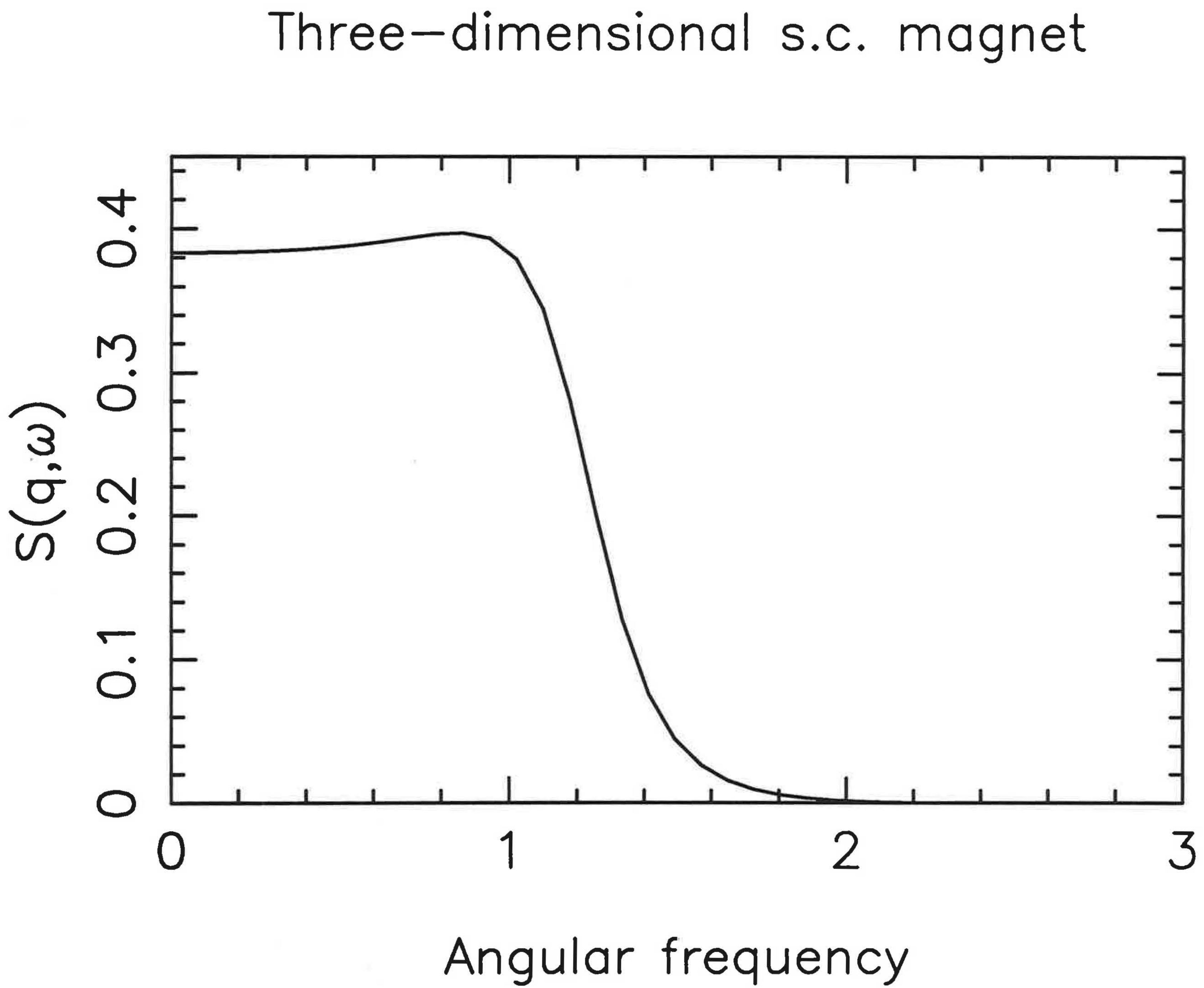


3b

Three-dimensional s.c. magnet



4a


Three−dimensional s.c. magnet
S(q,ω)
0
0.1
0.2
0.3
0.4
0
1
2
3
Angular frequency

(4b)