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Time-dependent spin-autocorrelation functions at $T = \infty$ for classical Heisenberg magnets in dimensionalities $d = 1$, 2, and 3 are investigated by means of a computer simulation. These functions are shown to exhibit power-law long-time tails of form $t^{-\alpha_d}$ with characteristic exponents α_d which differ significantly from the values $\alpha_d^{(SD)} = d/2$ predicted by the phenomenological spin-diffusion theory: $\alpha_1 = 0.609 \pm 0.005$, $\alpha_2 = 1.050 \pm 0.025$, $\alpha_3 \simeq 1.6$. The method to employ computer simulations for this problem differs from methods previously employed. Anomalous spin diffusion is confirmed by existing proton spin-lattice relaxation data for the quasi-1D $s = \frac{5}{2}$ Heisenberg antiferromagnet $(CH_3)_4NMnCl_3$.

The question as to what extent the transport of spin excitations in isotropic Heisenberg magnets at high temperatures can be described in terms of the phenomenological concept of spin diffusion is an old one,[1] but there exists no microscopic theory which confirms with some rigor the predictions of spin-diffusion theory. Nevertheless, spin diffusion has played an important role in the analysis and interpretation of dynamical experiments (NMR spin-lattice relaxation,[2-4] electron-spin resonance[5]) on insulating magnetic compounds, particularly on quasi few-dimensional systems where the effects are most pronounced.

Consider the Heisenberg model

$$H = -J\sum_{\langle ij \rangle} S_i \cdot S_j \qquad (1)$$

for a magnetic insulator with nearest-neighbor interaction between localized magnetic moments on a d-dimensional hypercubic lattice with periodic boundary conditions. The variable S_j represents the effective spin of the unpaired electrons on the magnetic ion. In the context of the theoretical model (1), the S_j can be regarded as spin operators or as classical three-component unit vectors. The spin dynamics is well defined in both cases. The occurrence of spin diffusion in the Heisenberg model hinges on the fact that the total spin $S_T = \sum_i S_i$ is a constant of the motion and on the postulate (which relies on that conversation law) that at sufficiently high temperatures the time evolution of fluctuations of the total spin,

$$S(q,t) = \sum_j \exp(iq \cdot r_j) S_j(t),$$

is governed by a diffusion equation

$$\frac{\partial S(q,t)}{\partial t} = -Dq^2 S(q,t) \qquad (2)$$

in the hydrodynamic regime, i.e., for sufficiently long wavelengths. The diffusion constant D is expected to depend on temperature and, for $T < \infty$, on whether the exchange coupling in (1) is ferromagnetic ($J > 0$) or anti-ferromagnetic ($J < 0$). Whereas this phenomenological concept is clearly classical in spirit, its implications for the time-dependent correlation functions are expected to hold for both the classical and the quantum Heisenberg models alike. The time evolution according to (2) implies that the correlation function $C(q,t) \equiv \langle S(q,t) \cdot S(-q,0) \rangle$ decays exponentially in time as

$$C(q,t) \sim e^{-Dq^2 t} \qquad (3)$$

asymptotically for small q and large t. An important consequence of this characteristic diffusive behavior is that the autocorrelation function $C_0(t) \equiv \langle S_j(t) \cdot S_j(0) \rangle$ exhibits a distinctive power-law long-time tail,

$$C_0(t) \sim t^{-\alpha_d}, \qquad (4)$$

with a characteristic exponent $\alpha_d^{(SD)} = d/2$, which strongly depends on the dimensionality of the diffusion process. In 1D and 2D systems, this long-time tail, if indeed present, is in turn responsible for infrared divergences of the type $\sim \omega^{-1/2}$ (1D) and $\sim \ln(1/\omega)$ (2D) in the frequency-dependent autocorrelation function $\Phi_0(\omega) \equiv \int dt\, e^{i\omega t} C_0(t)$, which are directly amenable to experimental investigation, e.g., by measurement of the magnetic field dependence of NMR spin-lattice relaxation rates.[2-4] Since the important question of whether the $T = \infty$ spin-autocorrelation function does in fact show the characteristic diffusive long-time tail (4) has never been established beyond ambiguity in spite of efforts to that end,[6,7] further theoretical investigations are called for. However, a recent study of the 1D $s = \frac{1}{2}$ Heisenberg model by Roldan, McCoy, and Perk[8] has shed interesting new light onto the problem (which I shall discuss later on) and arrived at the conclusion that a diffusive $t^{-1/2}$ tail is at least compatible with rigorous bounds derived from a moment expansion of $C_0(t)$.

This Letter reports a novel approach to the performance of computer simulations for the calculation of time-dependent correlation functions of classical spin systems (the focus here is on autocorrelation functions at

$T = \infty$). Choose the kth set of initial conditions $\{S_i^{(k)}\}$ at random and integrate the equations of motion

$$\dot{S}_i = -S_i \times \partial H / \partial S_i$$

numerically on a time interval $[0, t_{max}]$. The autocorrelation function of the canonical ensemble at $T = \infty$ is then simulated by a combined site and ensemble average,

$$C_0(t) = \frac{1}{K} \sum_{k=1}^{K} \frac{1}{N} \sum_{i=1}^{N} S_i(t) \cdot S_i^{(k)},$$

i.e., an average over all N lattice sites and an average over K randomly chosen initial spin configurations. For finite K, $C_0(t)$ is a fluctuating quantity, and for finite N, $C_0(t)$ is subject to finite-size effects. The goal then is to choose N and K sufficiently large to make both of these undesired effects small. In previous computer simulations of spin dynamics, the strategy has been to emphasize large N (typically $N = 1000$–4000)[6,9] in order to eliminate finite-size effects. The use of large N has the additional benefit, so it is argued, that the site average alone will reduce the statistical fluctuations in $C_0(t)$ to a reasonable level. Ensemble averaging is then no longer deemed essential and is, in general, performed only to a very small extent (typically $K = 3$–6).[6–9]

The strategy of the alternative approach proposed here is to reduce the statistical fluctuations up front by extensive ensemble averaging and then to eliminate finite-size effects by analysis of precise data systematically for increasing N. Naturally, this method can be executed in practice only for relatively small systems, systems in which finite-size effects are likely to be prominent, perhaps uncontrollably large. While the study of critical phenomena or certain types of propagating modes may thus be out of reach, an opportunity presents itself in situations where the coherence length is very small or, in fact, zero as is the case in $T = \infty$ spin dynamics. At infinite temperature, local spin fluctuations spread very slowly and persist over appreciable times and distances only if a conservation law prohibits them from decaying. Under such circumstances, the most efficient computer simulation focuses on small systems, enphasizes ensemble averaging, and eliminates finite-size effects by systematically increasing N.

Consider the 1D Heisenberg model, a ring of N spins. Figure 1 shows the $T = \infty$ autocorrelation function $C_0(t)$ for time lags up to $Jt_{max} = 10$ of four different short chains ($N = 4$, 6, 8, and 10) along with the result for a longer chain ($N = 50$). The simulation uses a simple fourth-order Runge-Kutta integration with fixed time step $Jdt = 0.005$–0.025, which proved to guarantee sufficient accuracy for all results reported in this Letter. The ensemble averaging over many thousand random initial conditions reduces the statistical uncertainties sufficiently to allow for a systematic elimination of finite-size effects.

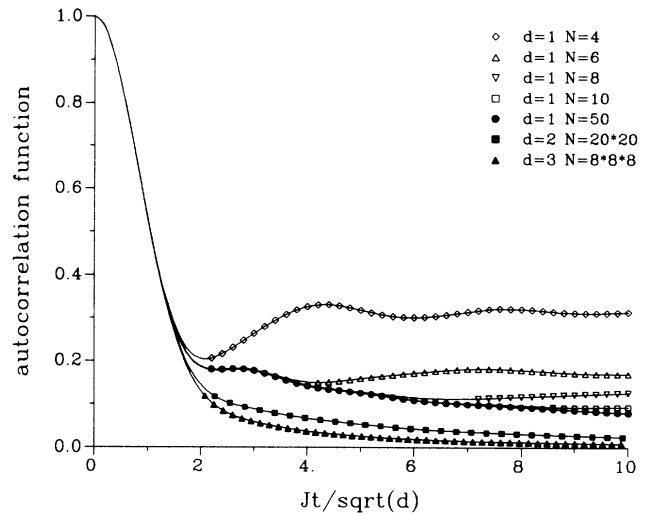For short time lags, all (1D) curves shown in Fig. 1 lie

FIG. 1. Spin-autocorrelation function $\langle S_i(t) \cdot S_1 \rangle$ of the classical Heisenberg model at $T = \infty$. The curves with open symbols represent simulation data averaged over $K = 20\,000$ random initial conditions for 1D systems of sizes $N = 4$, 6, 8, and 10. The curves with filled symbols show results for a linear chain ($N = 50$), a square lattice ($N = 20 \times 20$), and a simple cubic lattice ($N = 8 \times 8 \times 8$). They represent simulation data averaged over $K = 22\,792$ (1D), $K = 4563$ (2D), and $K = 4041$ (3D) random initial conditions.

on top of each other with great precision. The curve for the shortest chain ($N = 4$) is the first to deviate from the others (at $Jt \geq 1$). The curve representing the next longer chain ($N = 6$) starts to deviate from the remaining ones at some larger time lag ($Jt \geq 3$), etc. For $N \geq 14$, no noticeable finite-size effects occur on the time interval $[0,10]$. For all finite N, the function $C_0(t)$ decays to a positive constant at $t \to \infty$; that constant is a monotonically decreasing function of N which tends to zero as $N \to \infty$. The autocorrelation function for a chain of length $N = 50$ has been determined up to $Jt_{max} = 40$; there are no indications of finite-size effects on that interval. In fact, apart from the shallow minimum at $Jt \simeq 2.2$, the result shows hardly any structure in addition to the monotonic diffusive tail on that time interval (see discussion of Fig. 2 below).

Now consider the three curves in Fig. 1 which are decorated by filled symbols. They represent the autocorrelation functions for the 1D, the 2D (square lattice), and 3D (simple cubic lattice) Heisenberg models. The simulations were performed for lattices of sizes $N = 50$ (1D), $N = 20 \times 20$ (2D), and $N = 8 \times 8 \times 8$ (3D), respectively. It is considerably more difficult to avoid finite-size effects in more dimensions, particularly in 3D, without compromising on extensive ensemble averaging. Experience also reveals that finite-size effects in the 2D and 3D systems fade away somewhat less uniformly with increasing N than is the case in 1D systems. While this poses a problem for a detailed quantitative analysis, it
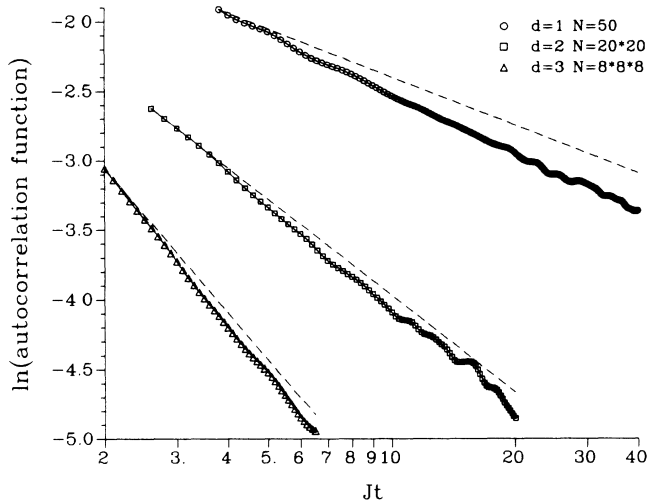
FIG. 2. Log-log plot of the autocorrelation functions $\langle S_i(t) \cdot S_i \rangle$ for the 1D, 2D, and 3D classical Heisenberg models from computer simulations (same data as also used in Fig. 1). The dashed lines represent the slopes, $-\alpha_d^{(SD)} = -d/2$, predicted by the phenomenological spin-diffusion theory.

does not obstruct the observation of the diffusive tail in the autocorrelation functions. Note that in Fig. 1 the time axis is rescaled by $\sqrt{d}$, which causes the three curves to superimpose very precisely for time lags somewhat beyond the inflection point. Here the diffusive long-time tail sets in, whose strength weakens with increasing d in qualitative agreement with the phenomenological spin-diffusion result.

A more detailed analysis of these diffusive tails, however, reveals that the simulation data show significant deviations from the predicted $t^{-d/2}$ behavior. This is illustrated in Fig. 2, which shows a doubly logarithmic plot of the 1D, 2D, and 3D autocorrelation functions over time intervals of different sizes. The data are in support of the spin-diffusion picture on a qualitative level: uniform power-law decay with a characteristic exponent which increases with increasing dimensionality. The weak nonuniformities of the 1D data on the time interval [20,40] are mostly due to residual statistical fluctuations. For increasing time lags, their elimination requires an increasing amount of ensemble averaging. The more pronounced nonuniformities of the 2D data on the time interval [10,20] are due to the onset of finite-size effects, which tend to make their initial appearance in the form of interferencelike ripples on top of an otherwise uniform power-law decay. Since such patterns are not expected to arise from a purely diffusive process, their systematic study promises to provide valuable insight into the underlying transport mechanism of spin fluctuations. The nonuniformities of the 3D data, finally, represent the ever-present oscillatory correction terms to the long-time tails, which have not yet decayed sufficiently on the time interval [2,6.5].

Comparison of the simulation data with the slopes of the phenomenological $t^{-d/2}$ prediction (represented by the dashed lines in Fig. 2) clearly demonstrates that spin diffusion in the classical Heisenberg magnet (1) is anomalous. The characteristic exponents α_d are larger than $d/2$ for all three cases. The deviation is largest (amounting to more than 20%) in the 1D system, for which the available simulation data are most extensive and precise. A quantitative analysis of the data shown in Fig. 2 and of data for systems of different sizes yields the following estimates for the characteristic exponents α_d:

$$\alpha_1 = 0.609 \pm 0.005, \quad \alpha_2 = 1.050 \pm 0.025.$$

For the 3D system, the data indicate that $\alpha_3 \simeq 1.6$, but it is uncertain to what extent the asymptotic behavior can be extracted from $\ln C_0(t)$ on the relatively short time interval [2,6.5].

For an interpretation of this anomalous spin diffusion observed in classical Heisenberg magnets, consider the Laplace transform of the time-dependent correlation function $C(q,t)$ in the continued-fraction representation:

$$\Gamma(q,z) \equiv \int_0^\infty dt \, e^{-zt} C(q,t)$$

$$= \cfrac{1}{z + \cfrac{\Delta_1(q)}{z + \cfrac{\Delta_2(q)}{z + \cdots}}}. \tag{5}$$

This representation results from Mori's projection-operator formalism and is constructed most conveniently by Lee's method of recurrence relations, which amounts to an orthogonal expansion of $S(q,t)$ in an abstract Hilbert space.[10] The $\Delta_n(q)$, which fully specify $\Gamma(q,z)$, are then expressible in terms of the norms of the orthogonal vectors f_n as $\Delta_n(q) = \|f_n\|^2 / \|f_{n-1}\|^2$, where

$$\|f_n\|^2 = Z_N^{-1} \int dS_1 \cdots dS_N e^{-\beta H} f_n f_n^*,$$

$Z_N = \int dS_1 \cdots dS_N e^{-\beta H}$, and $\beta = 1/k_B T$. The f_n are generated recursively from the relation $f_{n+1} = iLf_n + \Delta_n f_{n-1}$ with $f_{-1} = 0$, $f_0 = S(q,0)$, where $L = i[H,]$ is the Liouville operator expressed in the form of a Poisson bracket. The dominant long-time asymptotic behavior of the correlation function $C(q,t)$ is then governed by the singularity in $\Gamma(q,z)$ closest to the imaginary z axis. The spin-diffusion phenomenology now relies on the assumption that this dominant singularity is a pole at $Z = -Dq^2$ for small q which remains isolated in the limit $q \to 0$. In the 1D Heisenberg model, for example, we have $\Delta_1(q) = \frac{4}{3} J^2 (1 - \cos q) \simeq \frac{2}{3} J^2 q^2$. The phenomenological spin-diffusion assumption then implies that (5) can be replaced by

$$\Gamma_{SD}(q,z) = (z + Dq^2)^{-1},$$

$$D = \frac{2}{3} J^2 \lim_{\substack{q \to 0 \\ z \to 0}} \left[z + \cfrac{\Delta_2(q)}{z + \cdots} \right]^{-1},$$

without the alteration of the dominant singularity. $\Gamma_{SD}(q,z)$ is the Laplace transform of (3). However, the legitimacy of this representation is by no means guaranteed. In fact, the possibility was pointed out in Ref. 8 that in the limit $q \to 0$, the asymptotic behavior of $C(\mathbf{q},t)$ may be goverend by a confluence of singularities. This is likely to produce a long-time tail in the autocorrelation function with a characteristic exponent a_d which is anomalous. That is precisely what is observed in the present study.

While searching the literature for the physical realizations most suitable to investigate experimentally the occurrence of anomalous spin diffusion, I was surprised to discover that the phenomenon is, in fact, prominently present in three different NMR proton spin-lattice relaxation studies[2-4] on the quasi-1D $s = \frac{5}{2}$ Heisenberg antiferromagnet $(CH_3)_4NMnCl_3$ [tetramethyl ammonium manganese trichloride (TMMC)], but had remained unnoticed in the analysis at the time those studies were undertaken. Figure 5 of Ref. 2, Fig. 1 of Ref. 3, and Fig. 2 of Ref. 4 all show relaxation rates $1/T_1$ versus frequency or magnetic field,[11] all measured at room temperature ($k_BT/J \simeq 23$). The relaxation rate is expected to contain a frequency-independent contribution in addition to a contribution which is proportional to $\Phi_0(\omega)$. Hence, it should fit an expression of the form $1/T_1 = P\omega^{a_1-1} + Q$ with three variable parameters P, Q, and a_1.[2-4] Using two independent methods of nonlinear regression analysis, I have consistently obtained the following exponent values which best fit the three sets of experimental data, respectively:

$$a_1^{(\exp)} = 0.57 \pm 0.11 \quad (\text{Ref.2}),$$

$$a_1^{(\exp)} = 0.60 \pm 0.06 \quad (\text{Ref.3}),$$

$$a_1^{(\exp)} = 0.62 \pm 0.05 \quad (\text{Ref.4}).$$

In two out of three cases the value $a_1^{(SD)} = 0.5$ is clearly ruled out. These experimental results represent a striking confirmation of the occurrence of anomalous spin diffusion in the 1D $s = \frac{5}{2}$ Heisenberg antiferromagnet TMMC.

A more detailed account of this study including results for different model systems and a more explicit and exhaustive comparison with available experimental data will be published in due course. I wish to thank F. Borsa and J. P. Boucher for suggestions which were useful for my reanalysis of the TMMC data. This work was supported by the National Science Foundation, Grant No. DMR-86-03036 and by a grant from Research Corporation.

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