Nonequilibrium structure factor for conserved spin dynamics: Abrupt temperature increase

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We consider the nonequilibrium, elastic-scattering structure factor $S(q,t)$ (q denotes the wave vector, t the time), for the Kawasaki spin-conserving kinetic Ising model of a one-dimensional system with nearest-neighbor interactions, initially in equilibrium at temperature T_I, that is suddenly placed in contact with a heat bath at temperature T_F, with $T_F \gg T_I$. We present detailed results for the case of $T_F=\infty$, for which we have succeeded in calculating the exact form of $S(q,t)$. For finite T_F, we present an approximation scheme for the higher-order nonequilibrium correlation functions that leads to closure of the hierarchy of equations of motion. The merits of this approximation are that (i) $S(q,t)$ is guaranteed to satisfy an exact sum rule over the Brillouin zone (BZ) of wave vectors q, and (ii) $S(q,t)$ evolves to the correct value in the long-time limit. For antiferromagnetic coupling, the structure factor, initially dominated by the Bragg peak associated with T_I at the edge of the BZ, decays exponentially with time, e^{-t/τ_q} while approximately preserving its shape in q space, since the lifetime τ_q is nearly independent of q. Except near the center of the BZ, after the Bragg peak has decayed sufficiently, the dependence of $S(q,t)$ on q can be characterized as though the spins rapidly quasiequilibrate to the equilibrium structure factor associated with T_F, $\chi(q,T_F)$, in that $S(q,t)/\chi(q,T_F)$ is independent of q, but is time dependent, slowly approaching unity as $t^{-1/2}$ for large t. For $q\approx 0$ the initial form of S remains in effect until the value of t is of order q^{-2}. For ferromagnetic coupling, the initial Bragg peak for $q\approx 0$ does not preserve its shape while decaying exponentially, since the lifetime τ_q strongly depends on the wave-vector q, diverging as q^{-2} for $q\to 0$, and, in particular, it is as though the spins for $q\approx 0$ remain ''frozen'' at T_I. Analogous to the behavior for antiferromagnetic interactions, away from the center of the BZ, we find that $S(q,t)/\chi(q,T_F)$ is independent of q and is a function of t/t_w, very slowly approaching unity. The characteristic ''waiting time'' t_w is anomalously long, proportional to ξ^2, where ξ is the equilibrium correlation length at temperature T_I. This behavior of t_w can be related to the random walk of domain boundaries. [S1063-651X(96)00608-3]

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I. INTRODUCTION

The response of condensed-matter systems to rapid changes in external parameters (temperature, for example) is a challenging problem in nonequilibrium statistical mechanics [1–4]. A well-studied example [1–5] is the spinodal decomposition of a two-phase thermodynamic system subjected to a quench from a temperature T_I (above an ordering temperature T_C) to a temperature T_F (below T_C). As a result of the change in temperature, the initial system is no longer thermodynamically stable and subsequently evolves into domains of ordered phases as the system equilibrates at the lower temperature. The interest in such nonequilibrium systems arises from the fact that, in analogy with critical phenomena, the kinetics of domain formation for widely different systems can be classified according to a few universal growth laws, which depend only on conservation laws and on such factors as the number of ordered phases that can exist [4]. We note that, as a prefatory remark for the present work, the word ''nonequilibrium'' has two distinct connotations in the statistical physics literature. Nonequilibrium often refers to systems *slightly* removed from equilibrium by infinitesimal external influences; this is the well-known linear-response regime [6] where, through the fluctuation-dissipation theorem, the response of a system to first order in the external driving force is related to equilibrium-averaged time-correlation functions. Nonequilibrium, however, also refers to systems *strongly* out of equilibrium [1–4,7] where a large external perturbation is suddenly applied, thereby driving the system to a new configuration that is far removed from its initial equilibrium state. Relatively little is known about the subsequent time evolution of such systems toward eventual equilibrium, precisely because thermodynamic systems subject to strong perturbations do not fall within the linear-response regime. This article is concerned with one specialized model system driven strongly out of equilibrium for which an analysis of its time evolution can be performed.

A basic experimental probe of strongly nonequilibrium systems is the nonequilibrium elastic-scattering structure factor $S(q,t)$ where t is the time [1–5]. This quantity [see Eq. (2.8)] is the Fourier transform of the two-point, equal-time, order-parameter correlation function, just as for the familiar equilibrium structure factor, $\chi(q,T)$, but evaluated in a *nonequilibrium* ensemble. Thus $S(q,t)$ monitors the instantaneous internal structure of the system as it evolves in time from thermal equilibrium at T_I to that at T_F. During the growth of domains, for example, it is found that $S(q,t)$ obeys a time-dependent scaling relation $S(q,t)\propto F[qL(t)]$

where F is a scaling function and where $L(t)$ is a time-dependent length characterizing the average domain size [1–4]. The latter quantity is generally found to have a power-law time dependence $L(t) \approx t^{\tau}$ for sufficiently large t, and the exponent τ can be used to define universality classes.

The nonequilibrium structure factor $S(q,t)$ therefore summarizes a wide range of interesting physical phenomena, and, accordingly, there has been considerable effort devoted to calculating this fundamental quantity for various models. We note that such problems have been addressed through direct numerical simulations [8], with renormalization group methods [9], and in model calculations designed to emphasize either the short- or the long-time regimes [10]. However there appear to be very few model systems for which $S(q,t)$ can be evaluated *exactly* for all t. One model system for which the exact form of $S(q,t)$ has been derived [11] is the one-dimensional Glauber [12] spin-flip kinetic Ising model. We note that Ising spins $\sigma_i = \pm 1$, in spite of their simplicity, provide an adequate description of the equilibrium properties and phase diagrams of many systems, including systems of adsorbed particles. It is highly desirable, therefore, to develop kinetic Ising models of $S(q,t)$ to treat the strong nonequilibrium behavior of Ising-like systems.

In this article, we provide a second kinetic Ising model for which an exact expression for $S(q,t)$ can be derived. We calculate $S(q,t)$ for the one-dimensional (1D) Kawasaki spin-exchange kinetic Ising model [13,14] for a system with nearest-neighbor interactions, initially in equilibrium at temperature T_I, that is suddenly placed in contact with a heat bath at temperature T_F, with $T_F > T_I$. In this model the total number of particles in the system is conserved, and hence it can be used, in its higher-dimensional versions, to provide a description of, for example, the growth of ordered domains in binary alloys or in systems of adsorbed atoms. We show, among other results, that in the limit $T_F \to \infty$ one can derive the complete analytic expression for $S(q,t)$ without invoking any approximations. We also develop an approximate treatment for finite T_F. Before we discuss these results further, however, it will be useful to contrast the Kawasaki and Glauber models, which are the two most widely studied kinetic Ising models. In the Glauber model, the allowed dynamical transitions of the system are restricted to single spin flips. As a result, the Glauber model cannot describe hydrodynamic transport phenomena caused by long-range spatial inhomogeneities, e.g., diffusion, since the total spin, or, equivalently, the total number of particles in the system, is not a conserved quantity. The Kawasaki model, on the other hand, only allows for the simultaneous flip of two opposite nearest-neighbor spins, so that they ''exchange'' values. This mechanism does conserve the total spin and hence can be used to model transport phenomena. As we will see, the severe constraint imposed by this conservation law renders a theoretical treatment of $S(q,t)$ nontrivial, even for the case of a one-dimensional array of spins with T_F set equal to infinity.

Whereas Mazenko and Widom [11] were able to obtain $S(q,t)$ exactly for the one-dimensional Glauber model for arbitrary T_F, we are able to provide an exact solution for the Kawasaki model only for the special value $T_F = \infty$. This is due to the fundamentally different character of the dynamics of these two systems discussed above. As we will show, for arbitrary T_F the equation of motion for $S(q,t)$ in the one-dimensional Kawasaki model includes an infinite number of higher-order nonequilibrium correlation functions, besides the two-spin correlation functions in terms of which $S(q,t)$ is defined. By contrast, for the one-dimensional Glauber model the equation of motion for $S(q,t)$ contains only two-spin correlation functions for all T_F. Therefore to obtain $S(q,t)$ for the Kawasaki model, one would in principle have to solve an infinite hierarchy of coupled kinetic equations for the various correlation functions. When $T_F = \infty$, however, it happens that the hierarchy is explicitly truncated at the two-spin correlation function level, and, as it turns out, the resulting equation of motion for $S(q,t)$ can be solved without approximation. To the best of our knowledge, there is no other example in the literature of an exact solution for $S(q,t)$ in which the total number of particles in the system is conserved.

In the context of the Kawasaki model with $T_F = \infty$, the dynamics that drives the system to its final state corresponds, in the equivalent lattice-gas picture, to nearest-neighbor random hopping with double site occupancy excluded. Such dynamical problems have received considerable attention in their own right [15,16]. We note that while we explicitly consider an initial state corresponding to thermal equilibrium at temperature T_I, our method of solution is not restricted to this case and could be applied to the evolution of $S(q,t)$ for the disordering of an arbitrarily prepared initial state subject to random hopping dynamics. Specifically, given an arbitrary initial-state structure factor, $S(q,0)$, the subsequent evolution of $S(q,t)$ by nearest-neighbor hopping can be calculated exactly if $T_F = \infty$.

For the case of general T_F, we present an approximation scheme in Sec. III for the higher-order correlation functions that leads to closure of the hierarchy of equations of motion. We remark that the occurrence of infinite hierarchies of coupled equations is widespread in many body theory and that the associated issue of how to optimally terminate them is a delicate and subtle problem. In particular, the qualitative behavior of $S(q,t)$ can be very sensitive to the details of the truncation procedure. We note, then, that our truncation method preserves the following two important features: (i) $S(q,t)$ evolves to the correct value in the long-time limit and (ii) $S(q,t)$ obeys an exact sum rule over the wave vectors q of the Brillouin zone (BZ). Our major findings when we use our truncation method are as follows. For the case of *antiferromagnetic* coupling, if the spins are initially in equilibrium at a sufficiently low temperature so that $S(q,t)$ exhibits a strong Bragg peak for $q \approx \pm \pi/a$, where a is the lattice spacing, we find that the Bragg peak *initially* decays proportional to $\exp(-t/\tau_q)$ while *approximately preserving its shape* in q space. We also obtain an explicit expression for the lifetime τ_q; it is virtually independent of q for $q \approx \pm \pi/a$. We note that evidence for initial exponential decay of the Bragg peak, with approximate shape preservation, has been observed in experiment [17–19]. This is discussed further in Sec. V. We also show that if $\Omega(q)t \geq 4$, where $\Omega(q)$ is a wave-vector-dependent relaxation rate defined by (2.30), the dependence of $S(q,t)$ on q can be characterized as though the spins have essentially equilibrated to the equilibrium structure factor at the final temperature, $\chi(q,T_F)$. Specifically, when this inequality applies, the ratio $S(q,t)/\chi(q,T_F)$ is independent of q, but is time dependent and slowly ap-

proaches unity from above, with the correction term decaying with time as $t^{-1/2}$. For very small q, because $\Omega(q)$ vanishes like q^2, this regime applies only for ultralong times.

For *ferromagnetic* coupling between the spins, the Bragg peak for $q\approx 0$ persists for an enormous time period. The initial value, $\chi(q,T_I)$, although decaying as for antiferromagnetic interactions in an exponential manner e^{-t/τ_q}, the lifetime $\tau_q^{-1}=2\Omega(q)$ diverges as q^{-2} as $q\to 0$. This divergence is a direct consequence of the spin-conserving dynamics for the present model along with the sum rule obeyed by $S(q,t)$. In contrast to the result we obtain for antiferromagnetic interactions, because of the strong q dependence of τ_q, the Bragg peak for $q\approx 0$ does *not* preserve its shape in q space while decaying exponentially. Away from the center of the BZ, we find that $S(q,t)/\chi(q,T_F)$ is independent of q and is a function of t/t_w, which rises very slowly to unity. The characteristic ''waiting time'' t_w is anomalously long, proportional to ξ^2, where ξ is the equilibrium correlation length at temperature T_I. As shown in Sec. IV B 1, this behavior of t_w can be related to the random walk of domain boundaries.

It will be noted that for the problem considered here, the usual order of T_I and T_F considered in quench problems is reversed, i.e., we are interested in the time evolution of an initial equilibrium system after it is subject to a sudden *increase* in temperature. We are motivated by experiments on the disordering kinetics of initially ordered surface structures in Si(100) [18,19], where observations of the decay of the ''satellite peak'' in $S(q,t)$, corresponding to a loss of surface order, suggests a one-dimensional disordering process. A preliminary listing of some of the results presented in Sec. IV for the special case of $T_F=\infty$ have been reported in Ref. [18] without derivation.

The outline of this article is as follows. In Sec. II, we briefly review the equilibrium properties of Ising spins that will be of use, in particular, the equilibrium structure factor $\chi(q,T)$ and the sum rule it satisfies. We then define the nonequilibrium structure factor $S(q,t)$ and derive its exact equation of motion in the context of the Kawasaki spin-exchange model. The quantity $S(q,t)$ also satisfies a sum rule that will be of fundamental significance in our analysis. In Sec. III we present a truncation procedure that enables us to calculate $S(q,t)$ for any finite final temperature. In the context of this procedure it is shown that the time evolution of $S(q,t)$ is governed by the time dependence of the nonequilibrium, nearest-neighbor two-spin correlation function $G(t)$. This function satisfies an integral equation that we solve using Laplace transform techniques. In Sec. IV, we focus on the case where the spins are placed in contact with a heat reservoir at *infinite* temperature. For this case we can obtain the exact form of $S(q,t)$ without invoking the truncation procedure that we employ for finite T_F. We provide detailed numerical results for the evolution of $S(q,t)$ and give its asymptotic properties. Finally, in Sec. V we summarize our results and discuss issues for further study. Appendix A is devoted to establishing the properties of the dispersion integral $F(w,\tau)$ that plays a major role in the analysis of $S(q,t)$. In Appendix B we show that the methods developed in this article can readily be adapted to provide the exact form of $S(q,t)$ for the Glauber kinetic Ising model which does not possess a conserved variable. Once again the primary quantities of physical interest are expressible in terms of the dispersion integral $F(w,\tau)$.

II. FORMULATION OF MODEL

A. Equilibrium properties

We consider a one-dimensional lattice of N Ising spins $\sigma_i=\pm 1$. The equilibrium thermodynamic properties of this system are well known, and we briefly review some of the most salient results that will be used in later sections. Equilibrium averages are constructed with the probability distribution function,

$$P[\sigma]=Z^{-1}\exp(H[\sigma]), \tag{2.1}$$

where Z is the partition function, and where $H[\sigma]$ is the Ising nearest-neighbor Hamiltonian multiplied by $-\beta=-1/k_BT$,

$$H[\sigma]=K\sum_{i=1}^{N}\sigma_i\sigma_{i+1}\quad(\sigma_{N+1}\equiv\sigma_1) \tag{2.2}$$

with $K\equiv\beta J$ denoting the nearest-neighbor coupling constant, where J is the exchange interaction strength. Note that (anti)ferromagnetic spin couplings are implied by ($K<0$) $K>0$. It will be convenient to assume an infinite lattice ($N\to\infty$), for which the system has a critical point at zero temperature. For the nonequilibrium problem, to be discussed below, the final state of the system is associated with a coupling constant K_F, and we will study the evolution that results from the sudden change in coupling constant, $K=K_I\to K_F$, starting from an initial value K_I, with $|K_I|>|K_F|$.

In the following, we will require the two-spin equilibrium correlation function [20]:

$$\langle\sigma_i\sigma_j\rangle=[\tanh(K)]^{|i-j|}\equiv u^{|i-j|}, \tag{2.3}$$

where the brackets denote an average with respect to $P[\sigma]$ and where we have introduced the symbol $u\equiv\tanh(K)$. We see from (2.3) that the correlation length governing the exponential decay of the two-spin correlation function is given by $\xi^{-1}=\ln[\coth(|K|)]$. We will also require the Fourier spin transform $\sigma(q)$ which can serve as a general order parameter,

$$\sigma(q)=N^{-1/2}\sum_n\exp(iqna)\sigma_n, \tag{2.4}$$

where q is restricted to the one-dimensional BZ, $0\leqslant|q|\leqslant\pi/a$, and where a is the lattice spacing. (Henceforth we choose $a=1$.) Note that the ferromagnetic order parameter is recovered by considering the limit $q\to 0$, while the antiferromagnetic order parameter is obtained in the limit $q\to\pi$. This can readily be seen from the equilibrium elastic-scattering structure factor $\chi(q,T)$, which measures the spectrum of fluctuations in equilibrium at temperature T,

$$\chi(q,T)=\langle\sigma(-q)\sigma(q)\rangle. \tag{2.5}$$

Evaluation of $\chi(q,T)$ using (2.3) and (2.4) is straightforward, and in the limit $N\to\infty$ one obtains

$$\chi(q,T)=\frac{\sqrt{1-\gamma^2}}{1-\gamma\cos q}, \tag{2.6}$$

where $\gamma\equiv\tanh(2K)=2u/(1+u^2)$. For $K>0$, we have $\gamma\to1$ in the low-temperature limit and χ diverges for $q\to0$, i.e., the system is dominated by long-wavelength fluctuations. By contrast, for $K<0$, $\gamma\to-1$ for low temperatures, and χ diverges for $q\to\pi$, i.e., antiferromagnetic ordering develops in this limit. We see from (2.6) that χ is invariant under the transformation $K\to-K$ and $q\to\pi-q$. For $q=0$ and $q=\pi$, then, the peak value of χ is given by $e^{2|K|}$, whereas the corresponding minimum value at $q=\pi$ and $q=0$ is $e^{-2|K|}$.

The quantity $\chi(q,T)$ defined by (2.5) and given in (2.6) satisfies a temperature-independent sum rule,

$$\int_{-\pi}^{\pi}dq\ \chi(q,T)=2\pi. \tag{2.7}$$

This result is a direct consequence of the requirement that at each lattice site n, the Ising fixed-length spin condition $\sigma_n^2=1$ is satisfied. Henceforth, we will use the abbreviated notation $\chi(q)$ to denote the equilibrium structure factor (2.6).

B. Nonequilibrium structure factor

The dynamical response to a rapid change in temperature from T_I to T_F can be characterized by the nonequilibrium structure factor $S(q,t;T_I,T_F)$ which is defined as

$$S(q,t;T_I,T_F)=\sum_{\{\sigma\}}\sigma(-q)\sigma(q)P[\sigma,t]\equiv\langle\sigma(-q)\sigma(q)\rangle_t$$
$$=1+2\sum_{n=1}^{\infty}\langle\sigma_0\sigma_n\rangle_t\cos(nq), \tag{2.8}$$

where $P[\sigma,t]$ is a time-dependent probability distribution that depends on T_I and T_F and is specified below, and where the subscript on the angular brackets denotes a nonequilibrium ensemble average with respect to $P[\sigma,t]$ [21]. Note that (2.8) is simply the nonequilibrium generalization of the static structure factor defined by (2.5), i.e., it is the Fourier transform of the equal-time, two-spin correlation function evaluated in a nonequilibrium ensemble. This quantity can be measured in real-time elastic-scattering experiments after a rapid change in external parameters, typically by monitoring the evolution of a Bragg peak [1–4]. An important constraint on our analysis is that a sum rule analogous to (2.7) holds for $S(q,t;T_I,T_F)$, i.e.,

$$\int_{-\pi}^{\pi}dq\ S(q,t;T_I,T_F)=2\pi. \tag{2.9}$$

This too is a consequence of the Ising fixed-length spin condition. Note that (2.9) holds for all times. Henceforth, we will generally abbreviate our notation and denote the nonequilibrium structure factor by $S(q,t)$.

We remark that $S(q,t)$ should not be confused with the dynamic structure factor, which is the *equilibrium*-averaged, *inelastic*-scattering structure factor, which we denote by $C(q,t,T)$. That structure factor is the Fourier transform of the two-spin time-correlation function,

$$C(q,t,T)=\langle\sigma(-q,0)\sigma(q,t)\rangle$$
$$=C_0(t,T)+2\sum_{n=1}^{\infty}C_n(t,T)\cos(nq), \tag{2.10}$$

where $C_n(t,T)\equiv\langle\sigma_0(0)\sigma_n(t)\rangle$ is the correlation of a spin at lattice site 0 at time $t=0$ with a spin at site n at time t evaluated in an *equilibrium* ensemble. In the following, we will simplify our notation and suppress the variable T, i.e., we write $C(q,t)$ and $C_n(t)$. We will calculate and discuss $C(q,t)$ in Sec. II D because of its heuristic value in anticipating results for $S(q,t)$ that are given in later sections.

The first task we face, in order to derive $S(q,t)$, is to establish the form of the nonequilibrium probability distribution function $P[\sigma,t]$. In kinetic Ising models this quantity is taken to satisfy a Markovian master equation [14], which, in operator form, we denote by

$$\frac{\partial}{\partial t}P[\sigma,t]=\sum_{\{\sigma'\}}D[\sigma|\sigma']P[\sigma',t]\equiv D_\sigma P[\sigma,t], \tag{2.11}$$

where the operator D_σ contains the typical gain and loss transition rates of a master equation, in this case assumed to arise from the interactions between the spin system and a heat reservoir. Once D_σ is specified, $P[\sigma,t]$ is formally given by [22]

$$P[\sigma,t]=\exp(D_\sigma t)P_I[\sigma], \tag{2.12}$$

where we have chosen the initial condition $P[\sigma,t=0]=P_I[\sigma]$, with $P_I[\sigma]$ the equilibrium distribution characterized by the initial-state coupling constant K_I. The quantity D_σ is a matrix operator in the 2^N-dimensional space of spin configurations, and is constructed so as to exhibit the specified spin dynamics, in our case a nearest-neighbor spin exchange, as well as to satisfy the requirement that $P[\sigma,t]$ evolves to the correct long-time limit, the equilibrium distribution $P_F[\sigma]$ characterized by the final-state coupling constant K_F. The latter requirement is fulfilled by constructing D_σ so that it satisfies detailed balance about the final-state equilibrium, i.e., $D[\sigma|\sigma']P_F[\sigma']=D[\sigma'|\sigma]P_F[\sigma]$, which is sufficient to show that P_F remains stationary, i.e., $D_\sigma P_F[\sigma]=0$. Note that in the most straightforward implementation of detailed balance, D_σ is a function only of K_F and is independent of K_I. In the next subsection we derive an explicit analytic representation of D_σ for the one-dimensional Kawasaki kinetic Ising model.

Combining (2.8) and (2.12), we obtain a formal equation of motion for $S(q,t)$,

$$\frac{\partial}{\partial t}S(q,t)=\sum_{\{\sigma\}}\sigma(-q)\sigma(q)D_\sigma P[\sigma,t]$$
$$\equiv\langle\widetilde{D}_\sigma[\sigma(-q)\sigma(q)]\rangle_t, \tag{2.13}$$

where $\widetilde{D}_\sigma$ is the adjoint of D_σ, with matrix elements $\widetilde{D}[\sigma|\sigma']\equiv D[\sigma'|\sigma]$. The quantity $\widetilde{D}_\sigma$ is the effective time-derivative operator for observables (i.e., spin functions) [23], whereas D_σ operates on distribution functions. The complete expression for (2.13) is fairly involved and is derived in the next subsection. In Sec. III we discuss the solution to (2.13), subject to the constraint (2.9).

C. Equation of motion

In this subsection we review those elements of the theory of kinetic Ising models that are necessary to derive the equation of motion for $S(q,t)$, Eq. (2.13). The reader uninterested in the details should proceed to (2.28).

As mentioned above, kinetic Ising models are defined by assuming that $P[\sigma,t]$ satisfies a Markov master equation. Master equations have the general ''gain-loss'' form [24]

$$\frac{\partial P[\sigma,t]}{\partial t}=\sum_{\{\sigma'\}}\left(W[\sigma|\sigma']P[\sigma',t]-W[\sigma'|\sigma]P[\sigma,t]\right)$$
$$=\sum_{\{\sigma'\}}\left(W[\sigma|\sigma']-\delta_{\sigma,\sigma'}\sum_{\{\sigma''\}}W[\sigma''|\sigma']\right)P[\sigma',t]$$
$$\equiv\sum_{\{\sigma'\}}D[\sigma|\sigma']P[\sigma',t]\equiv D_\sigma P[\sigma,t], \tag{2.14}$$

where the quantities $W[\sigma|\sigma']$ give the probability per unit time for a transition from the state of the system $\{\sigma'\}$ to the state $\{\sigma\}$. In the Markov approximation, the transition probabilities are functions only of the current configuration of the spins, and not the history of the system. For the Kawasaki spin-exchange model [14], the allowed transitions are restricted to the simultaneous flip (''exchange'') of two spins of opposite values. This ensures that the total spin is conserved during transitions. We can therefore write the basic form of the master equation operator [25,26] as

$$D[\sigma|\sigma']=\tfrac{1}{2}\sum_{i,j}\delta^{[i,j]}_{\sigma,\sigma'}D^{i,j}[\sigma|\sigma'], \tag{2.15}$$

where the sum is over pairs of spins i,j, and the quantity

$$\delta^{[i,j]}_{\sigma,\sigma'}=\prod_{k\neq i,j}\frac{1}{2}\left(1+\sigma_k\sigma'_k\right) \tag{2.16}$$

ensures that all spins *except* those at sites i and j remain unchanged. The operator D_σ therefore describes the stochastic exchange of all pairs of spins i and j while the rest of the spins remain momentarily fixed. From the form of (2.14), we can write the operator describing the exchange of spins i and j as

$$D^{i,j}[\sigma|\sigma']=W_{i,j}(\sigma')(\delta_{\sigma_i,\sigma'_j}\delta_{\sigma_j,\sigma'_i}-\delta_{\sigma_i,\sigma'_i}\delta_{\sigma_j,\sigma'_j}), \tag{2.17}$$

where the first set of δ functions ensures the spin-exchange mechanism $\sigma'_j\to\sigma_i$ and $\sigma'_i\to\sigma_j$, and $W_{i,j}(\sigma')$ is the probability per unit time for the exchange. Using the spin representation of the Kronecker function $\delta_{\sigma_i,\sigma'_j}=\frac{1}{2}(1+\sigma_i\sigma'_j)$ we obtain the explicit expression,

$$D^{i,j}[\sigma|\sigma']=-\tfrac{1}{4}W_{i,j}(\sigma')(\sigma_i-\sigma_j)(\sigma'_i-\sigma'_j). \tag{2.18}$$

Restricting ourselves now to nearest-neighbor spin exchange, the form of the master equation operator is given by

$$D[\sigma|\sigma']=-\tfrac{1}{8}\sum_i\sum_{\nu=\pm1}W_{i,i+\nu}(\sigma')\delta^{[i,i+\nu]}_{\sigma,\sigma'}$$
$$\times(\sigma_i-\sigma_{i+\nu})(\sigma'_i-\sigma'_{i+\nu}), \tag{2.19}$$

where the sum on ν is over nearest neighbors. This expression corrects that given in Ref. [26], which differs by an overall factor of two. An essential requirement for any master equation is that the normalization of the probability distribution be conserved in time; from (2.14) this is satisfied if $\sum_{\{\sigma\}}D[\sigma|\sigma']=0$. This basic requirement is readily seen to be satisfied by (2.19).

We have yet to specify the transition probability function W. The simplest form of W for nearest-neighbor spin exchange in one dimension is that due to Zwerger [27]

$$W_{i,i+\nu}(\sigma)=\alpha\left[1-\frac{1}{2}\gamma_F(\sigma_{i-\nu}\sigma_i+\sigma_{i+\nu}\sigma_{i+2\nu})\right], \tag{2.20}$$

where $\gamma_F=\tanh(2K_F)$, and α is an overall frequency setting the spin-exchange rate for uncoupled spins and is taken as a phenomenological parameter of the model. In Sec. II D, it is shown that α can be identified as the spin-diffusion coefficient in the high-temperature limit. Equation (2.20) is a sufficient, but not necessary, condition to guarantee that detailed balance is satisfied in the final-state equilibrium. Detailed balance does not uniquely determine the form of W and hence there is some freedom in choosing this function: It can be multiplied by any spin function that is independent of σ_i and $\sigma_{i+\nu}$ and still satisfy detailed balance. Therefore, depending on the precise form of W, classes of generalized kinetic Ising models can be defined that are consistent with the basic constraint of detailed balance [28]. As shown by Mazenko and Oguz [29], the Zwerger form (2.20) results in the least complicated nonlinearities in the single-spin equation of motion, which is our reason for adopting this form. Note, however, that results for $S(q,t)$ derived for $K_F=0$ do not depend on the functional form of W. In this limit, W reduces to a constant transition rate, *independent* of the local spin configurations, which, we note, corresponds to unbiased random hopping in the associated lattice-gas picture.

To evaluate the equation of motion for $S(q,t)$ (2.13) one must first have the basic dynamical equations obeyed by Ising spins for spin-exchange kinetics. Using (2.19), and the definition of the adjoint operator $\widetilde{D}[\sigma|\sigma']\equiv D[\sigma'|\sigma]$ [see discussion around (2.13)], the equation of motion for a single spin can be derived,

$$\widetilde{D}_\sigma\sigma_i=-\sum_{\nu=\pm1}W_{i,i+\nu}(\sigma)(\sigma_i-\sigma_{i+\nu}). \tag{2.21}$$

The interpretation of (2.21) as the time rate of change for a single spin is transparent: Spin exchange is blocked when nearest-neighbor spins are in the same state; when exchange can occur, it does so with a probability determined through detailed balance by the difference in energy environments that would result from the exchange. Note that by summing (2.21) over all lattice sites, the total spin is manifestly conserved for all temperatures.

Besides the single-spin equation of motion, (2.21), we will also require the equation of motion for the product of two spins. Using (2.19), the following can be derived:

$$\widetilde{D}_\sigma \sigma_i \sigma_j = \sigma_i \widetilde{D}_\sigma \sigma_j + \sigma_j \widetilde{D}_\sigma \sigma_i + 2 \sum_{\nu=\pm 1} (\delta_{j,i} - \delta_{j,i+\nu}) W_{i,i+\nu}(\sigma)(1 - \sigma_i \sigma_{i+\nu}). \tag{2.22}$$

The last group of terms in (2.22) serve the following purposes. If $j=i$, the result should be zero since $\sigma_i^2=1$; this is guaranteed by the $\delta_{j,i}$ term. The $\delta_{j,i+\nu}$ term reflects a locally conserved quantity under the exchange process: The product of two nearest-neighbor spins should remain invariant to internal permutation. The $\delta_{j,i+\nu}$ term insures that the product of nearest-neighbor spins evolves only through exchange with the ''outside'' neighbors. We note that these seemingly innocuous terms play an extremely important role in the theory. If either or both of these terms were absent from the equation of motion (2.22), $S(q,t)$ would not evolve to the correct steady-state solution, namely, the equilibrium structure factor evaluated at the final temperature $\chi(q,T_F)$.

We can now derive the equation of motion for $S(q,t)$. By substituting (2.20) in (2.21), the explicit single-spin equation of motion is given by

$$\widetilde{D}_\sigma \sigma_i = -\alpha \Big\{ 2\sigma_i - \sigma_{i-1} - \sigma_{i+1} + \frac{1}{2}\gamma_F(\sigma_{i-2} - \sigma_{i-1} - \sigma_{i+1} + \sigma_{i+2}) + \frac{1}{2}\gamma_F(2\sigma_{i-1}\sigma_i\sigma_{i+1} - \sigma_{i-2}\sigma_{i-1}\sigma_i - \sigma_i\sigma_{i+1}\sigma_{i+2}) \Big\}, \tag{2.23}$$

which we note contains nonlinear, three-spin terms in addition to single-spin terms. Upon Fourier transforming (2.23), we have

$$\widetilde{D}_\sigma \sigma(q) = -\omega(q)\sigma(q) - 2\alpha\gamma_F \sin^2(q/2) V(q), \tag{2.24}$$

where $\omega(q)$ is a temperature- and wave-vector-dependent frequency,

$$\omega(q) = 2\alpha \sin^2(q/2)[2(1-\gamma_F \cos q) - \gamma_F], \tag{2.25}$$

and where $V(q)$ is the Fourier transform of the three-spin term,

$$V(q) \equiv N^{-1/2} \sum_n \exp(iqn)\sigma_{n-1}\sigma_n\sigma_{n+1}. \tag{2.26}$$

We note that if we had employed a transition probability function other than (2.20), additional nonlinear terms [29] besides $V(q)$ would occur in (2.24). Note that the right-hand side of (2.24) decreases continuously to zero as $q \to 0$, independent of the temperature. This is a direct manifestation of the conservation law. Now, upon Fourier transforming (2.22) we obtain,

$$\widetilde{D}_\sigma \sigma(-q)\sigma(q) = 2\sigma(-q)\widetilde{D}_\sigma \sigma(q) + \frac{2}{N}\sum_{i,\nu}[1 - \exp(iqv)] W_{i,i+\nu}(\sigma) \times (1 - \sigma_i \sigma_{i+\nu}), \tag{2.27}$$

which is what is required in (2.13). We remark that the form of the spin-exchange operator (2.19) is general and describes the Kawasaki spin dynamics on an arbitrary dimensional lattice, for any choice of the transition probability function W. Equations (2.21), (2.22), and (2.27), which are derived using (2.19), are therefore also general equations of motion for the nearest-neighbor Kawasaki kinetic Ising model, irrespective of the dimensionality of the lattice and the form of W. For our one-dimensional model, combining (2.24) and (2.27) together with the Zwerger transition probability (2.20), we obtain the desired equation of motion,

$$\frac{\partial S(q,t)}{\partial t} = -2\omega(q)S(q,t) - 4\alpha\gamma_F \sin^2(q/2)\langle \sigma(-q)V(q)\rangle_t + 8\alpha \sin^2(q/2)[1 - (1+\gamma_F)\Phi_1(t) + \gamma_F \Phi_2(t)], \tag{2.28}$$

where $\Phi_m(t) \equiv \langle \sigma_0 \sigma_m \rangle_t$ for $m=1,2$ are the nonequilibrium nearest-neighbor and next-nearest-neighbor correlation functions, respectively.

We see from (2.28) that the equation of motion for $S(q,t)$ generates a complicated four-spin nonequilibrium correlation function $\langle \sigma(-q)V(q)\rangle_t$. We stress that this term represents an infinite sum of four-spin correlations, as can be seen from the identity,

$$\langle \sigma(-q)V(q)\rangle_t = \Phi_2(t) + 2\sum_{n=1}^{\infty} \langle \sigma_{-1}\sigma_0\sigma_1\sigma_n\rangle_t \cos(nq). \tag{2.29}$$

In principle, therefore, to solve (2.28) one would have to obtain an equation of motion for general four-spin products, which, in turn, would couple to six-spin terms. In general, one would have to solve an *infinite hierarchy* of coupled kinetic equations for the various nonequilibrium correlation functions. We note that this is in contrast to the equation of motion obeyed by $S(q,t)$ for Glauber dynamics (see Appendix B) which involves *only* two-spin correlation functions. To make progress, then, some means of truncating the hierarchy must be found. A specific proposal is presented and implemented in Sec. III.

For the special case of infinite final temperature $K_F=0$ an *exact* solution for $S(q,t)$ becomes possible since the hierarchy is explicitly terminated, i.e., (2.28) will involve only two-spin terms. The form of $S(q,t)$ in this special case is discussed in Sec. IV. Note that in this limit ($K_F=0$), the single-spin equation of motion (2.23) reduces to the usual equation of motion for random hopping dynamics [15,16].

Returning to the general case ($K_F \neq 0$), it will be useful to rewrite the equation of motion (2.28) in a form in which it can be explicitly seen that $S(q,t)$ approaches the correct long-time limit, and in addition, formally satisfies the sum

rule (2.9). We first introduce a new characteristic frequency $4\alpha\Omega(q)$ where the dimensionless quantity $\Omega(q)$ is defined as

$$\Omega(q)\equiv(1-\gamma_F\cos q)\sin^2(q/2), \tag{2.30}$$

so that $\omega(q)=4\alpha\Omega(q)-2\alpha\gamma\sin^2(q/2)$ in (2.25). Then, utilizing (2.8) and (2.29), together with (2.30), and the following result that holds in equilibrium:

$$\Omega(q)\chi(q,T_F)=\sin^2(q/2)\left[1+\frac{\gamma_F}{2}-(1+\gamma_F)\mathbf{\Phi}_1(\infty)+\frac{\gamma_F}{2}\Phi_2(\infty)\right], \tag{2.31}$$

where $\Phi_m(\infty)=u_F^m$, it is easy to show from (2.28) that the equation of motion is given by

$$\frac{\partial S(q,t)}{\partial t}=-8\alpha\sin^2(q/2)[(1-\gamma_F\cos q)\{S(q,t)-\chi(q,T_F)\}+\gamma_F W(q,t)+G(t)]. \tag{2.32}$$

The quantities $W(q,t)$ and $G(t)$ are defined as

$$W(q,t)\equiv\sum_{n=2}^{\infty}[\langle\sigma_{-1}\sigma_0\sigma_1\sigma_n\rangle_t-\langle\sigma_0\sigma_n\rangle_t]\cos(nq), \tag{2.33}$$

and

$$G(t)\equiv(1+\gamma_F)[\Phi_1(t)-u_F]-\frac{\gamma_F}{2}[\Phi_2(t)-u_F^2]. \tag{2.34}$$

We first see from (2.32) that $S(q,t)$ will evolve to the correct long-time limit only if the quantity $W(q,t)$ vanishes in equilibrium $W(q,\infty)=0$. This does indeed occur because of a special property of equilibrium correlation functions for the one-dimensional Ising model with nearest-neighbor interactions and zero magnetic field [30],

$$\langle\sigma_{-1}\sigma_0\sigma_1\sigma_n\rangle=\langle\sigma_0\sigma_n\rangle\quad(|n|\geqslant 1). \tag{2.35}$$

Thus the terms in (2.33) all vanish in the long-time limit, i.e., $W(q,\infty)=0$. We note therefore that any proposed approximation for the four-spin *nonequilibrium* correlation functions, e.g., for truncating the hierarchy discussed above, must preserve this property, i.e., that the differences $\langle\sigma_{-1}\sigma_0\sigma_1\sigma_n\rangle_t-\langle\sigma_0\sigma_n\rangle_t$ vanish in the long-time limit, in order for the approximate $S(q,t)$ to evolve to the correct long-time value.

We also see that the infinite series in (2.33) starts with the term $n=2$, because the $n=1$ term vanishes identically. This has an important consequence. By integrating over all wavevectors q in the BZ, it is easy to check that the sum rule (2.9) is formally obeyed by (2.32), as well as by (2.28). The advantage of (2.32), however, is that the sum rule will be satisfied *independent* of the values of the terms in $W(q,t)$; this is true only because $W(q,t)$ excludes the $n=1$ term. This is an important theoretical result. It guarantees that, no matter what approximation we devise for the four-spin correlation functions, the ensuing approximate result for $S(q,t)$ will satisfy the same sum rule (2.9) as does the exact solution.

Finally, we note that by writing $\omega(q)=4\alpha\Omega(q)-2\alpha\gamma\sin^2(q/2)$ in (2.28) we can group together the terms in the equation of motion in a way that reveals some important features that would not have been readily transparent otherwise. The quantity $\omega(q)$ cannot properly be interpreted as a relaxation rate, since, as seen from (2.25), it becomes negative for various combinations of temperature and wave vector, namely, whenever $2(1-\gamma_F\cos q)<\gamma_F$. This seeming occurrence of an instability is specious, however, and underscores that the single-spin function $\sigma(q)$ is not an eigenmode of the dynamics, as can be seen from (2.24). Moreover, the higher-order spin correlation functions play an important role in providing overall stability, since it is known [31] that if the operator $\tilde{D}_\sigma$ satisfies detailed balance, its eigenvalues are all real and negative (or nonpositive) and hence the evolution of $S(q,t)$ is bounded for all times. On the other hand, the quantity $\Omega(q)$ defined by (2.30) is positive for all temperatures and wave vectors, and thus it properly serves as a relaxation rate.

Equations (2.32)–(2.34) provide the exact equation of motion for $S(q,t)$ in the Kawasaki-Zwerger model. These equations serve as the starting point for the truncation proposal we present in Sec. III. Before proceeding to that proposal, however, we devote the following subsection to a heuristic discussion of the expected long-time behavior of $S(q,t)$ using the asymptotic properties of the equilibrium time-correlation functions.

D. Asymptotic analysis of time-correlation functions

To have a better understanding of the results given in Secs. III and IV for the nonequilibrium structure factor $S(q,t)$ it will be useful to examine at an arbitrary temperature the asymptotic properties of the time-correlation functions of equilibrium fluctuations produced by the spin-exchange dynamics. This is because, for the nonequilibrium system, in the asymptotic approach to equilibrium, one would intuitively expect a close relationship between the long-time form of nonequilibrium ensemble averages and the time-correlation functions of spontaneous fluctuations about equilibrium. Indeed, this is the qualitative content of the Onsager regression hypothesis [6,32]. We therefore expect that the long-time form of the strongly nonequilibrium response will coincide with the *asymptotic* time dependence predicted by linear-response theory, i.e., by the dynamics of equilibrium fluctuations. Information about the dynamics of equilibrium fluctuations can be extracted from the dynamic structure factor $C(q,t)$ given in (2.10), and we will examine this quantity in this subsection. We remark, however, that the properties of the equilibrium time-correlation functions can *only* yield insight into the asymptotic form of the nonequilibrium response; an analysis of the dynamics of equilibrium fluctuations clearly cannot serve as a substitute for solving the nonequilibrium problem. For example, the results of this subsection cannot yield information about the time required after the sudden change in the heat bath for the onset of the asymptotic approach to equilibrium.

The asymptotic decay of the equilibrium time-correlation functions $C_n(t)$ in (2.10) can be established if we know the form of $C(q,t)$, since, using (2.10), we have

$$C_n(t)=\frac{1}{2\pi}\int_{-\pi}^{\pi}dq\ \exp(-inq)C(q,t). \qquad (2.36)$$

Now $C(q,t)$ can be most easily obtained by considering the Laplace transform

$$\widetilde{C}(q,s)=\int_0^\infty dt\ \exp(-st)C(q,t)\equiv L_t[C(q,t)]. \qquad (2.37)$$

In the memory function representation [6], the transform $\widetilde{C}(q,s)$ is written as

$$\widetilde{C}(q,s)=\frac{\chi(q)}{s+\varphi(q,s)}. \qquad (2.38)$$

The quantity $\varphi(q,s)$, the memory function, is a generalized space- and time-dependent relaxation rate and contains all the information about the dynamics of the system in the linear-response regime. In general, $\varphi(q,s)$ is a complicated object involving higher-order dynamical correlation functions; an explicit expression is given in Ref. [33]. In particular, $\varphi(q,s)$ cannot be obtained in closed form for this model, which is a direct consequence of the infinite hierarchy discussed in Sec. II C. We can, however, rigorously establish the form of the memory function as $q\to 0$, which is what we require to find the asymptotic properties of the time-dependent correlation functions $C_n(t)$. First, we know that $\varphi(q,s)$ will vanish as q^2 as $q\to 0$ because of the conservation law. Hence we can write that $\varphi(q,s)=q^2D(q,s)$ where $D(q,s)$ is nonzero as $q\to 0$. For small q, then, the Laplace transform $\widetilde{C}(q,s)$ has a ''hydrodynamic'' pole at $s=-q^2D(0,0)+O(q^4)$ provided that $D(0,s)$ is well behaved as $s\to 0$. As shown by Mazenko and Oguz [29], however, for the Zwerger model $D(q,s)$ is *independent* of s for small q. We may therefore set $D(0,0)=D$, where D is the diffusion coefficient,

$$D\equiv\lim_{s\to 0}\lim_{q\to 0}q^{-2}\varphi(q,s)=\alpha(1-u)(1-\gamma), \qquad (2.39)$$

where we have given the exact value of D that results for this model [27,29]. Note that the parameter α can be identified as the diffusion coefficient in the high-temperature limit. By taking the inverse Laplace transform of $\widetilde{C}(q,s)$ we thus arrive at the key result, that for small q and all times t, the leading form of the structure factor $C(q,t)$ is given by,

$$C(q,t)\sim\chi(q)\exp(-Dq^2t)\quad(q\to 0). \qquad (2.40)$$

We note that this is precisely the form that $C(q,t)$ would adopt had we started with the continuum diffusion equation, instead of the lattice-based spin-exchange kinetic Ising model. In the following we will refer to an approximate, ''hydrodynamic'' structure factor, $C^H(q,t)$, which we define to have the form (2.40) throughout the BZ, as well as hydrodynamic correlation functions, $C_n^H(t)$ defined by using (2.36) in conjunction with $C^H(q,t)$. We will show that in many cases this approximation is remarkably accurate.

We first examine the simplest case of infinite temperature for which $\chi(q,T=\infty)=1$ for all q, and $D=\alpha$; we consider the case of general T below. That $\chi(q)$ is independent of q expresses the fact that in the high-temperature limit there is no permanent structure to the system at any length scale. Dynamically, however, $C(q,t)$ measures the temporal correlation of spatially separated fluctuations. As we will see, that there is a nontrivial time dependence to the spin correlations in this noninteracting limit is a reflection of the conservation law, which in essence enforces a *dynamic* correlation between spatially separated spins. This dynamic coupling effect can clearly be seen from the correlation functions $C_n(t)$ which we examine first in the hydrodynamic approximation.

It is convenient to introduce a local time variable, appropriate to the lattice site n, defined by $\tau_n\equiv 2\alpha t/n^2$. Using (2.36) and the definition of $C^H(q,t)$ we have

$$nC_n^H(t)=\frac{1}{\pi}\int_0^{n\pi}du\ \exp\left(-\frac{1}{2}u^2\tau_n\right)\cos u. \qquad (2.41)$$

If $\tau_n n^2\geqslant 2$ we may extend the upper limit to infinity so that we obtain

$$nC_n^H(t)=(2\pi\tau_n)^{-1/2}\exp[-1/(2\tau_n)],\quad \tau_n n^2\geqslant 2. \qquad (2.42)$$

This result states that the quantities $nC_n^H(t)$, if plotted versus the local time variable τ_n, will be given by a single universal curve as long as $\tau_n n^2\geqslant 2$. This approximate scaling formula is a lattice analog of the *exact* scaling property satisfied by the solutions to the usual diffusion equation (which is the continuum limit of a lattice random walk) for a δ-function initial condition, $C(r,t)=(4\pi Dt)^{-1/2}\exp(-r^2/4Dt)$.

It is of interest to compare the result of (2.42), which we obtained within the hydrodynamic approximation, with the exact result for $nC_n(t)$ for the special case of $T=\infty$. In this limit we can obtain the exact memory function $\varphi(q,s)=4\alpha\sin^2(q/2)$ since in this case $\sigma(q)$ is an eigenmode of the dynamics [see (2.24)] [34]. We then have the exact result for all q and all t,

$$C(q,t)=\exp[-4\alpha\ \sin^2(q/2)t]\quad(T=\infty) \qquad (2.43)$$

which, we note, is consistent with (2.40) for small q. Combining (2.36) and (2.43), we have the exact expression,

$$C_n(t)=\frac{1}{2\pi}\int_{-\pi}^{\pi}dq\ \exp[-4\alpha t\ \sin^2(q/2)-iqn]. \qquad (2.44)$$

This integral is readily evaluated to yield

$$C_n(t)=\exp(-2\alpha t)I_n(2\alpha t)\quad(T=\infty), \qquad (2.45)$$

where I_n is a modified Bessel function. An analysis of the integral representation (2.44) shows that for $\tau_n n^2\geqslant 2$, this equation reduces to the scaling formula (2.42) that we obtained in the hydrodynamic approximation. Note also, from (2.45), that $C_n(0)=\delta_{n,0}$, as should be the case in the high-temperature limit. We can see, however, that starting at $t=0$, spatially separated spins *develop* correlations because of the spin-conserving dynamics. This dynamic correlation effect is evident in Fig. 1.

The correlation function (2.45) will be recognized as the probability distribution for a one-dimensional continuous-time random walk that starts at the origin [15]. This is not surprising given the connection between the Kawasaki model

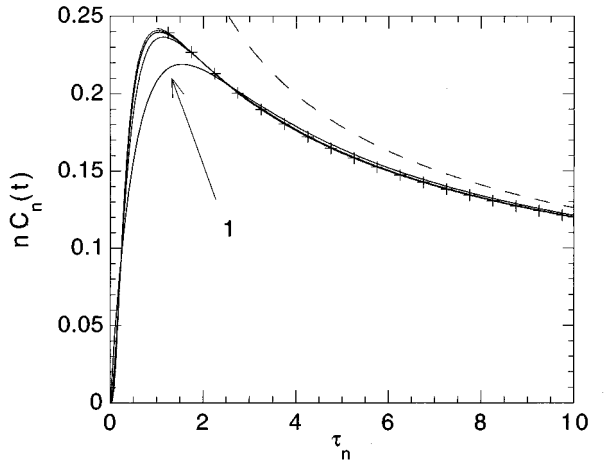

FIG. 1. Product of the lattice-site index n with the time-correlation functions $C_n(t)$, given by (2.45), as a function of the dimensionless local time variable $\tau_n \equiv 2\alpha t/n^2$. The arrow identifies the curve for $n=1$, while those for $n=2$, 3, and 4 are nested successively. The curves for $n \geqslant 2$ and $\tau_n > 2$ are very well described by the universal curve (2.42), shown as + symbols. The dashed curve shows the leading asymptotic term, (2.46), for $nC_n(t)$ that follows from (2.42) or (2.45). The correlations are maximized at approximately $\tau_n \approx 1$, confirming the picture that the spins interact by a random walk process.

at $T=\infty$ and nearest-neighbor random hopping. Thus two spins that interact by a random walk process, initially separated by n lattice sites, should be most strongly correlated after a characteristic time proportional to n^2. We can thus expect that all of the correlation functions C_n should have a maximum for virtually the same value of the local time variable $\tau_n = 2\alpha t/n^2$. In fact, Montroll [35] showed that, for large n, (2.45) is maximized when the value of the independent variable $2\alpha t$ is given by $n^2 + 1/2 + O(1/n)$, i.e., when, $\tau_n \sim 1 + 1/(2n^2) + O(1/n^3)$. All of these properties are consistent with our earlier remark that C_n is most appropriately considered as a function of a local time variable, appropriate to the lattice site n.

We display in Fig. 1 the quantities $nC_n(t)$ [obtained from (2.45)] plotted against the local time variable τ_n. Also shown are the results obtained from (2.42), denoted by the + symbols. It is quite remarkable that the hydrodynamic result is in effect, at least for $n \geqslant 2$, for such early times. We also have, from either (2.45) and the asymptotic properties of Bessel functions, or from (2.42), that

$$nC_n(t) \sim (2\pi\tau_n)^{-1/2} \quad (\tau_n \gg 1, \quad T=\infty). \tag{2.46}$$

This quantity is shown in Fig. 1 as the dashed curve.

For an arbitrary finite value of the temperature, the analysis of $C_n(t)$ is considerably more complicated and we will present only the asymptotic form of $C_n(t)$ for long times, the analog of (2.46). For long times only the small-q regime contributes to (2.36). It is then legitimate to extend the limits of integration in (2.36) to $\pm\infty$, use (2.40), and replace $\chi(q)$ by $\chi(0)$. We thus obtain,

$$C_n(t) \sim \frac{1}{2\pi} \int_{-\infty}^{\infty} dq \ \chi(q) \exp[-Dq^2 t - iqn]$$

$$\sim \frac{\chi(0)}{\sqrt{4\pi D t}}, \quad (Dt \gg L^2), \tag{2.47}$$

where L is a length defined as follows. For ferromagnetic interactions, $L = \max(n, \xi)$, i.e., $L = \xi, n$ for $n <, > \xi$. Here ξ is the equilibrium correlation length, given by $\xi^{-1} = \ln(\coth K) \sim \exp(-2K)$ as $K \to \infty$. For antiferromagnetic interactions, however, $L = n$. We see that for sufficiently long times, $C_n(t)$ decays with a $t^{-1/2}$ power-law form, a result that is solely a consequence of the conservation law in one dimension. Note that the time required before the onset of the $t^{-1/2}$ regime depends on the temperature as well as on n. From (2.39), for ferromagnetic interactions and for low temperatures, D vanishes as $D \sim \alpha\xi^{-3}$ and hence as $\xi \to \infty$ the ''waiting time'' obtained from (2.47) $D^{-1}\xi^2$ diverges [36] as ξ^5. For antiferromagnetic interactions, $D \to 4\alpha$ for low temperatures, and one attains the $t^{-1/2}$ regime relatively rapidly.

The main result of this subsection is that at all temperatures, for sufficiently long times $Dt \gg L^2$ the equilibrium time-correlation functions $C_n(t)$ are described by the $t^{-1/2}$ power-law form, a result that follows directly from the dynamics that features a conserved mode. As discussed at the beginning of this subsection, this leads us to expect similar behavior for the nonequilibrium structure factor $S(q,t)$ in the long-time limit. Specifically, in the asymptotic approach to equilibrium, we expect that $S(q,t)$ will also be characterized by diffusive behavior, the signature of which is a $t^{-1/2}$ time dependence in one dimension. This asymptotic power-law form is indeed found in the results derived below. We will find however that the time required to *attain* the asymptotic regime depends strongly on the initial temperature T_I and the sign of the coupling constant.

III. DETERMINATION OF $S(q,t)$

A. Preliminaries

In Sec. II C we derived the equation of motion (2.32) satisfied by the nonequilibrium structure factor $S(q,t)$ for the present model upon assuming that the system is suddenly exposed to a heat bath at a temperature T_F. In the remainder of this article the dimensionless quantity $4\alpha t$ will be denoted by t so that the equation of motion reads

$$\frac{\partial S(q,t)}{\partial t} = -2\sin^2(q/2)[(1-\gamma_F \cos q)\{S(q,t) - \chi(q,T_F)\}$$

$$+ G(t) + \gamma_F W(q,t)], \tag{3.1}$$

where $W(q,t)$ and $G(t)$ are given by (2.33) and (2.34), respectively. We remind the reader that W and G depend on T_I and T_F, as well as the arguments explicitly listed. As discussed in Sec. II C, the integral $\int_{-\pi}^{\pi} dq \ W(q,t)\sin^2(q/2)$ vanishes for all times. Thus, if we integrate (3.1) on q over the BZ, the integral $\int_{-\pi}^{\pi} dq \ S(q,t)$ will be time-independent as long as we require that

$$G(t) = -\frac{1}{\pi} \int_{-\pi}^{\pi} dq' [S(q',t) - \chi(q',T_F)] \Omega(q'), \tag{3.2}$$

where $\Omega(q)$ is defined by (2.30). That is, any solution of (3.1) which also satisfies (3.2) will at all times satisfy the sum rule (2.9), so long as the proposed initial form for $S(q,0)$ satisfies that rule. Also note that if one combines (3.1) and (3.2), the rate of change $\partial S(q,t)/\partial t$ depends not only on the current value of $S(q,t)$, but also on the current value of $S(q',t)$ for all other wave vectors q' within the BZ. Put differently, the role of the sum rule (2.9) is to cause the equation of motion to be highly nonlocal in q space, even though it is local in time (i.e., Markovian).

In Sec. II C we also pointed out that the four-spin nonequilibrium correlation function, $\langle \sigma_{-1}\sigma_0\sigma_1\sigma_n\rangle_t$, which appears in the definition (2.33) of $W(q,t)$, satisfies an equation of motion involving still other, higher-spin nonequilibrium functions. In particular, (3.1) is but the first of an infinite hierarchy of equations of motion. For the present model, the only case where one can avoid this hierarchy without invoking a truncation procedure is when the final temperature T_F is infinitely large, so that the parameter γ_F vanishes. In particular, the forms of $\langle \sigma_{-1}\sigma_0\sigma_1\sigma_n\rangle_t$ and $\Phi_2(t)$ are irrelevant when $T_F=\infty$ and (3.1) reduces to

$$\frac{\partial S(q,t)}{\partial t} = -2\sin^2(q/2)[S(q,t)-1+G(t)] \quad (T_F=\infty). \tag{3.3}$$

In Sec. IV we provide a detailed analysis of (3.3) and (3.2) leading to the exact analytical form of both $S(q,t)$ and $G(t)=\Phi_1(t)$ for this special case.

In the next subsection we present an approximation procedure for arbitrary *finite* T_F that truncates the hierarchy at the level of nonequilibrium two-spin correlation functions. We summarize here the major results that emerge upon assuming that the spins are initially in equilibrium. For the case of antiferromagnetic interactions, if T_I is sufficiently low, the initial peak in $S(q,t)$ at first decays exponentially while approximately retaining its original shape. As time proceeds, this gives way to the following. In the regime $\Omega(q)t>4$, $S(q,t)$ exhibits ''quasiequilibrated'' behavior $S(q,t)/\chi(q,T_F)\approx A(t)$ independent of q, where $A(t)$ evolves towards unity with a power-law $t^{-1/2}$ decay. In the vicinity of the center of the BZ, $S(q,t)$ retains its original (numerically, very small) form for extremely long times, until $t>O(q^{-2})$. For the case of ferromagnetic interactions, the initial peak near $q\approx 0$ also decays exponentially but with a decay time proportional to q^{-2}. The original shape of the peak is thus not preserved as time proceeds due to the conservation law that forces $S(0,t)$ to remain constant. In the regime $\Omega(q)t>4$, $S(q,t)$ also exhibits quasiequilibrated behavior $S(q,t)/\chi(q,T_F)\approx F(t/t_w)$ except that the time is scaled in units of a ''waiting time'' t_w. These issues are explored in detail in Sec. III C.

B. Truncation procedure for finite T_F

With the exception of a handful of special cases (e.g., the 1D Glauber model; see Appendix B), all theories of the nonequilibrium structure factor must confront the issue of truncating an infinite hierarchy of equations of motion, and some approaches have proven successful (as an example, the Langer, Bar-on, and Miller theory [37]). We note, however, that the qualitative behavior of the ensuing approximate equations of motion can be extremely sensitive to the details of the truncation scheme. Thus, the long-time steady-state solution may be incorrect, or the approximate theory may only be valid for long wavelengths. For the present system, an exact treatment for an arbitrary finite final temperature appears to be hopelessly intractable. It is therefore of interest to develop an approximation for $W(q,t)$ that is rigorously valid whenever the system is in thermal equilibrium, i.e., for $t=0$ and for $t=\infty$. This will be sufficient to ensure that the result for $S(q,t)$, obtained using the approximate theory, will in fact evolve to the correct long-time value, namely, $S(q,t)\rightarrow\chi(q,T_F)$ as $t\rightarrow\infty$.

We replace the four-spin function $\langle \sigma_{-1}\sigma_0\sigma_1\sigma_n\rangle_t$ in (2.33) by the two-spin function $\langle \sigma_0\sigma_n\rangle_t$. That is, we impose on the nonequilibrium four-spin function the same equality with the nonequilibrium two-spin function as applies [see (2.35)] for the thermal equilibrium versions of these spin functions for any temperature for the one-dimensional nearest-neighbor Ising model. As a consequence of this procedure the function $W(q,t)$ vanishes identically and (3.1) reduces to

$$\frac{\partial S(q,t)}{\partial t} = -2\sin^2(q/2)[(1-\gamma_F\cos q)\{S(q,t)-\chi(q,T_F)\} + G(t)]. \tag{3.4}$$

We stress that while (3.4) is in general an approximate equation of motion, it is rigorously valid at $t=0$ and at $t=\infty$ for arbitrary finite T_F. It is also rigorously valid for all times when $T_F=\infty$. In essence, our truncation procedure consists of constraining the time evolution of the four-spin nonequilibrium correlation function *as if* this function at any moment of time is characterized by a common, global time-dependent temperature [38] that ultimately reaches the final value T_F. We remark that $W(q,t)$ is not identically zero for $T_F=\infty$; only because $\gamma_F=0$ is (3.4) rigorously valid in the high-temperature limit. In the next paragraph, however, we give arguments that $W(q,t)\equiv 0$ should be a good approximation for this system.

We have already remarked that the approximation $W(q,t)\equiv 0$ is compatible with both $S(q,t)$ achieving its correct long-time value, and the sum rule (2.9) being satisfied. In this paragraph we provide arguments that even in an exact treatment $W(q,t)$ is not expected to contribute appreciably to the evolution of $S(q,t)$ [39]. We note that our basic approximation, $\langle \sigma_{-1}\sigma_0\sigma_1\sigma_n\rangle_t\approx\langle \sigma_0\sigma_n\rangle_t$ will, for all times and temperatures, become progressively more accurate as n increases. Only for relatively small values of $|n|$ should the correlation of σ_n with σ_0 differ *appreciably* from the correlation of σ_n with the local group of spins $\sigma_{-1}\sigma_0\sigma_1$. Hence, in the function $W(q,t)$, which is the Fourier transform of the *differences* $\langle \sigma_{-1}\sigma_0\sigma_1\sigma_n\rangle_t-\langle \sigma_0\sigma_n\rangle_t$, [see (2.33)], we expect the terms in the summation to decay rapidly after some characteristic value of n. For this reason, we do not expect $W(q,t)$ to ever become large, as for example compared with $S(q,t)$. Furthermore, for a given value of n, this approximation will also become more accurate the lower the temperature, since in this case the product of spins $\sigma_{-1}\sigma_0\sigma_1$ will effectively have the same value as the spin σ_0 for both ferro- and antiferromagnetic couplings. Thus, for low temperatures,

we expect that the effective number of terms in (2.33) to achieve convergence of $W(q,t)$ will be *reduced* over that for higher temperatures. This is in direct contrast to the behavior of $S(q,t)$, where, because long-range correlations are induced at low temperatures, progressively more and more terms must be included in (2.8) to achieve convergence, and we obtain the strong peaks in this function for $q=0$ and $q=\pi$. This implies, then, that the *same* circumstances that lead to strong peaks in $S(q,t)$ will also result in $W(q,t)$ being relatively small for all wave vectors. Hence, we expect that $W(q,t)$ starting from its *exact* initial value $W(q,0)=0$, will remain small compared to $S(q,t)$ before it eventually decays to zero for long times. We therefore believe that $W(q,t)=0$ (for all times) is an excellent approximation for this system.

Our task now reduces to solving the pair of Eqs. (3.4) and (3.2). The procedure we invoke consists of solving (3.4) for $S(q,t)$, treating $G(t)$ as a given function, and then imposing (3.2) upon the solution. Following this procedure we find that $G(t)$ satisfies the Volterra integral equation (3.7) given below. The kernel of that integral equation is of convolution form,and thus we will be able to find [see Sec. III D] the exact solution by invoking Laplace transform methods. We now turn to the details.

The formal solution of (3.4) is given by

$$S(q,t)=S(q,0)e^{-2\Omega(q)t}+\chi(q,T_F)(1-e^{-2\Omega(q)t})-J(q,t), \tag{3.5}$$

where

$$J(q,t)\equiv 2\sin^2(q/2)\int_0^t dt'\ G(t')\exp[-2\Omega(q)(t-t')]. \tag{3.6}$$

In writing (3.5), the initial form of S has been left arbitrary, other than satisfying the obligatory sum rule (2.9). Substituting (3.5) in (3.2), one finds that the unknown function $G(t)$ satisfies a Volterra integral equation of the first kind [40,41]

$$\int_0^t d\tau\ P(t-\tau)G(\tau)=Q(t), \tag{3.7}$$

where the kernel is given by

$$P(t)=2\int_{-\pi}^{\pi} dq\ \sin^2(q/2)\exp[-2\Omega(q)t], \tag{3.8}$$

and

$$Q(t)=\int_{-\pi}^{\pi} dq[S(q,0)-\chi(q,T_F)]\exp[-2\Omega(q)t]. \tag{3.9}$$

The initial and final forms of S appear in the function $Q(t)$, and we note that $Q(0)=0$ as a consequence of (2.9). We use Laplace transforms in Sec. III D to solve (3.7) for $G(t)$ as well as to obtain an expression for $J(q,t)$ using (3.6).

To assess the significance of the term $J(q,t)$ in (3.5), it is useful to invoke one of the standard mean-value theorems [42] for integrals. Given an integral of the form $\int_a^b dt\ g(t)h(t)$, where in the closed interval $[a,b]$ both g and h are continuous and h does not change sign, then $\int_a^b dt\ g(t)h(t)=g(x_M)\int_a^b dt\ h(t)$ for at least one value x_M in $[a,b]$. We may thus rewrite (3.6) as

$$\begin{aligned} J(q,t)&=2\sin^2(q/2)G(\xi_q t)\int_0^t dt'\exp[-2\Omega(q)(t-t')]\\ &=G(\xi_q t)(1-\gamma_F\cos q)^{-1}\{1-\exp(-2\Omega(q)t]\}, \end{aligned} \tag{3.10}$$

where ξ_q is a number in the interval $[0,1]$ that depends on t as well as the value of q. Using (2.6) and (3.10), we arrive at the following exact, alternate representation of (3.5),

$$\begin{aligned} S(q,t)=S(q,0)e^{-2\Omega(q)t}&+\chi(q,T_F)[1-G(\xi_q t)(1-\gamma_F^2)^{-1/2}]\\ &\times(1-e^{-2\Omega(q)t}). \end{aligned} \tag{3.11}$$

Generally, in the remainder of this section we suppose that the system is initially in thermal equilibrium at temperature T_I, so that in (3.11) we may make the replacement $S(q,0)=\chi(q,T_I)$. Thus in the next subsection we will focus our attention on

$$\begin{aligned} S(q,t)=\chi(q,T_I)e^{-2\Omega(q)t}&+\chi(q,T_F)\\ &\times[1-G(\xi_q t)(1-\gamma_F^2)^{-1/2}](1-e^{-2\Omega(q)t}). \end{aligned} \tag{3.11$'$}$$

We will also suppose that $T_I \ll T_F$, and in particular that $|K_I|=|J|/(k_B T_I)\geq 2$ and $|K_F|=|J|/(k_B T_F)\leq 0.5$. Thus, for ferromagnetic (antiferromagnetic) coupling between spins, we have $\chi(q,T_I)\gg\chi(q,T_F)$ for $q\approx 0$ ($q\approx\pm\pi$), whereas in the remainder of the BZ we have $\chi(q,T_I)\ll\chi(q,T_F)$.

C. General properties of *S*

In the following we establish some general properties of $S(q,t)$ that can be inferred even prior to deriving the detailed form of $G(t)$ and $J(q,t)$. Only in Sec. III D do we provide a detailed derivation of those functions.

1. Exponential time decay of initial correlations

Consider first the case of antiferromagnetic interactions and wave-vectors $|q|\approx\pi$. Note that for such q we can replace $\Omega(q)$ by $\Omega(\pi)=1+\gamma_F$ in the first term of (3.11$'$). The large peak in S for $|q|\approx\pi$ initially decays exponentially with time, with a time constant $\tau^{-1}=2(1+\gamma_F)$ independent of q. The first term of (3.11$'$) can thus play a dominant role over a lengthy time interval measured by when that term has decreased sufficiently so as to become of the same order of magnitude as the second term. We also note that for these values of q, and for the time interval just described, as $S(q,t)$ decreases with time, *its initial shape* $S(q,0)=\chi(q,T_I)$ *is preserved*. Specifically, a semilog plot of $S(q,t)/\chi(q,T_I)$ will exhibit a linear dependence on t with slope $2(1+\gamma_F)$, *independent* of q for wave vectors sufficiently close to the BZ boundaries. We will discuss this result in Sec. IV for the case $T_F=\infty$ and in Sec. V for general temperatures, especially with regard to experimental evidence [18,19] for this behavior.

By contrast, for ferromagnetic interactions, at $t=0$ we have $S(q,0)=\chi(q,T_I)$, featuring a sharp peak for $q\approx 0$. Con-

sidering a small, fixed value of $|q|$ featuring $\chi(q,T_I)\gg 1$, we note from (3.11′) that as t increases from zero, the time dependence of S features exponential decay, with a time constant given by $\tau_q^{-1}=2\Omega(q)$. Indeed, one can essentially ignore the second term of (3.11′) for a large multiple of τ_q, until such a time that the first term has decreased to become of the same order as $\chi(q,T_F)$. Clearly τ_q diverges as q^{-2} for $q\to 0$. Because of this strong dependence on q there is no shape preservation of the decaying peak, as occurs for the case of antiferromagnetic interactions.

Binder [43] has given heuristic arguments that, if the final state is in a disordered phase and for wave vectors in the hydrodynamical regime, $S(q,t)$ should maintain exponential decay behavior even for $t\to\infty$. Such a form, however, i.e., (3.5) with $J(q,t)$ identically zero, would be correct only if $G(t)$ itself were identically zero, in contradiction to the integral equation (3.7) which is a direct consequence of the sum rule (2.9). That is, *exponential decay in the long-time limit cannot occur for the spin-conserving kinetic Ising model and still satisfy the sum rule*. We anticipate that a similar statement applies for higher-dimensional systems. The asymptotic behavior of S for large times is treated in Sec. III D 2. We show that $J(q,t)$ decreases to zero for long times as $t^{-1/2}$; however, the onset time for this behavior depends on q and grows as q^{-2} for $q\to 0$. Hence, for any nonzero value of q, at sufficiently long times the term $J(q,t)$ will greatly dominate over the decaying exponential in (3.5). Nevertheless, exponential time dependence does provide an accurate description of the time evolution of the structure factor during the first stage, and as we have seen, the duration of this stage increases monotonically with decreasing q, growing as q^{-2} for $q\to 0$.

2. Quasiequilibrated spins [$\Omega(q)t\geq 4$]

The result (3.5) for $S(q,t)$ calls for the evaluation of the function $J(q,t)$ defined by (3.6), and in turn, the function $G(t)$ which is the solution of the integral equation (3.7). The subsequent development, centered on the mean-value theorem, led to the results (3.11) and (3.11′), which are fully equivalent to (3.5). The form of (3.11) hints at the possibility that some of the major qualitative properties of $S(q,t)$ can be established even at this stage prior to the full calculation of $J(q,t)$ and $G(t)$. We base this remark on the fact that a knowledge of the q and t dependence of the parameter ξ_q is fully equivalent to possessing the function $J(q,t)$, yet this parameter lies in the range [0,1] and is surely a continuous function of its variables. This hint is confirmed in the following.

We show that ξ_q gradually increases with t and approaches unity from below, and more specifically, $G(\xi_q t)\approx G(t)$ if $\Omega(q)t\geq 4$. This greatly simplifies (3.11′) and allows us to arrive at an important general result, even prior to obtaining the explicit form of $G(t)$. It should be noted that the regime $\Omega(q)t\geq 4$ commences at a relatively early time $t\approx 4/(1+\gamma_F)$ for wave vectors near the edge of the BZ and for increasingly later times deeper within the zone. For values of q in the immediate vicinity of the center of the BZ where $\Omega(q)$ is vanishingly small, the requirement $\Omega(q)t\geq 4$ will be met only at extremely late times. In the case of ferromagnetic interactions, when $\Omega(q)t\geq 4$ we may discard the first term of (3.11′) because the numerical value of $\chi(q,T_I)$ is negligible except for $q\approx 0$. For the case of antiferromagnetic interactions we must retain the first term of (3.11′) despite the smallness of $e^{-2\Omega(q)t}$ since $\chi(q,T_I)$ is very large for $|q|\approx\pi$ and thus this term can dwarf the second term over an extended time interval.

In the regime $\Omega(q)t\geq 4$, the factor $e^{-2\Omega(q)(t-t')}$ in (3.6) is very small except in the immediate vicinity of the upper limit, namely, $t'\approx t$. We may therefore replace the function $G(t')$ by $G(t)$, with the result

$$J(q,t)\approx 2\sin^2(q/2)G(t)\int_0^t dt'\exp[-2\Omega(q)(t-t')]$$

$$\approx G(t)(1-\gamma_F\cos q)^{-1}\quad[\Omega(q)t\geq 4]. \tag{3.12}$$

Comparing with (3.10) we then have that

$$G(\xi_q t)\approx G(t)\quad[\Omega(q)t\geq 4] \tag{3.13}$$

so that, as claimed, the parameter ξ_q increases with time and $\xi_q\approx 1$ for $\Omega(q)t\geq 4$. An immediate consequence of (3.13) is that we may legitimately approximate the exact result (3.11′) by

$$S(q,t)\approx\chi(q,T_I)e^{-2\Omega(q)t}+[1-G(t)(1-\gamma_F^2)^{-1/2}]\chi(q,T_F)$$

$$[\Omega(q)t\geq 4]. \tag{3.14}$$

The interesting feature of (3.14) is the second term whose dependence on q bears the signature of the heat bath at the temperature T_F. *We may say, once $\Omega(q)t\geq 4$, that the spins have essentially equilibrated to their final temperature*. There is a multiplicative time-dependent amplitude $[1-G(t)(1-\gamma_F^2)^{-1/2}]$ that very gradually approaches unity for long times. This factor will be smaller (larger) than unity for ferromagnetic (antiferromagnetic) interactions. This can readily be seen as a direct manifestation of the underlying spin-conserving dynamics for this model. For values of q in the immediate vicinity of the center of the BZ where $\Omega(q)$ is vanishingly small, the requirement $\Omega(q)t\geq 4$ will be met only at extremely late times. Thus, for the antiferromagnet, to compensate for the ''hole'' in the values of S for very small values of $|q|$, which persists for an enormously long-time period, and yet to satisfy the sum rule (2.9), the above amplitude factor must be larger than unity, i.e., $G<0$. On the other hand, for the ferromagnet where initially S is very large for small values of $|q|$, to satisfy the sum rule, the amplitude must be smaller than unity, i.e., $G>0$. We will find in Sec. III D 2 by an asymptotic analysis of the solution of the integral equation (3.7), that S decreases as $t^{-1/2}$ for long times. The power-law exponent $-1/2$ is due to the fact that for small q, the condition $\Omega(q)t\geq 4$ is equivalent to $q\geq At^{-1/2}$ where A is a constant.

The key result of this subsection is that at late times the time dependence of $S(q,t)$ is governed by that of the short-range correlation functions contained in $G(t)$.

3. The regime $S(q,t)/\chi(q,T_F)\ll 1$

Using (2.6) one may rewrite the equation of motion (3.4) as

$$\frac{\partial S(q,t)}{\partial t} = 2\sin^2(q/2)\{(1-\gamma_F^2)^{1/2}[1-S(q,t)/\chi(q,T_F)] - G(t)\}. \quad (3.15)$$

Now, in a regime where $S(q,t)/\chi(q,T_F) \ll 1$ we may integrate (3.15) so as to obtain the approximate relation

$$S(q,t) \approx \chi(q,T_I) + 2\sin^2(q/2)\left[(1-\gamma_F^2)^{1/2}t - \int_0^t dt' G(t')\right]. \quad (3.16)$$

Inspecting this result we may conclude that (3.16) should apply in the regime where both $\chi(q,T_I)/\chi(q,T_F)$ and $t\sin^2(q/2)$ are small compared to unity. In that regime the departure of $S(q,t)$ from $\chi(q,T_I)$ is proportional to $\sin^2(q/2)$, with a time-dependent proportionality constant. For antiferromagnetic interactions this behavior will apply even for relatively long times for values of $|q|$ in the immediate center of the BZ. For the case of ferromagnetic interactions, the condition $S(q,t)/\chi(q,T_F) \ll 1$ will apply for values of q which are not near the center of the BZ yet fulfill the condition that $t\sin^2(q/2)$ be small compared to unity. The applicable range of q values rapidly shrinks with time.

D. Determination of $S(q,t)$

1. Solution of integral equation (3.8)

In the preceding subsection we were able to arrive at a number of significant conclusions concerning $S(q,t)$ without establishing the detailed properties of $G(t)$ and $J(q,t)$. In this subsection we determine both of these functions by providing the formal solution of the integral equation (3.7) for an arbitrary choice of the initial function $S(q,0)$ and any value of T_F. The left side of (3.7) is a convolution integral, so that $g(s)=q(s)/p(s)$, where g, p, and q denote the Laplace transforms of G, P, and Q, respectively, defined by (2.37). Now

$$q(s) = \int_{-\pi}^{\pi} dq[S(q,0)-\chi(q,T_F)][s+2\Omega(q)]^{-1}, \quad (3.17)$$

and

$$p(s) = 2\int_{-\pi}^{\pi} dq\ \sin^2(q/2)[s+2\Omega(q)]^{-1}. \quad (3.18)$$

Thus,

$$G(t) = L_t^{-1}\left\{p^{-1}(s)\int_{-\pi}^{\pi} dq[S(q,0)-\chi(q,T_F)] \times [s+2\Omega(q)]^{-1}\right\}, \quad (3.19)$$

where L_t^{-1} denotes the inverse Laplace transform, which can be expressed in terms of the standard Bromwich contour [44] chosen to the right of all singularities of the integrand.

We will now show that (3.19) may be reduced to a simpler form (3.21) upon exploiting the analytic properties of $p(s)$ and $q(s)$. Both of these functions are expressed as dispersion integrals, so each is an analytic function of s, with the exception of a branch cut extending from the origin to $-2\Omega_{\max}$, where $\Omega_{\max}$ denotes the maximum value of the function $\Omega(q)$. Referring to (2.30), one finds that $\Omega_{\max}=1+\gamma_F$ for $\gamma_F \geq -1/3$, $\Omega_{\max}=(1+|\gamma_F|)^2/(8|\gamma_F|)$ for $-1\leq\gamma_F\leq -1/3$. Furthermore, because $S(q,0)$, $\chi(q,T_F)$, and $\Omega(q)$ are real quantities, we have that $[p(s)]^*=p(s^*)$, with a similar relation applying for $q(s)$, and thus also for $q(s)/p(s)$. Given these properties of p and q, the ratio $q(s)/p(s)$ is also analytic in s except for the branch cut just described; it would have isolated poles corresponding to zeros of $p(s)$, if such were to exist. However, $p(s)$ has no zeros. To show this we obtain an explicit expression for $p^{-1}(s)$ using the substitution $\cos q=(1-y)/(1+y)$ in (3.18) and Eq. (3.197.1) of Ref. [45], with the result

$$p^{-1}(s) = (4\pi)^{-1}[s+2(1+\gamma_F)]^{1/2}\{[s+1-\gamma_F-\Delta(s)]^{1/2} + [s+1-\gamma_F+\Delta(s)]^{1/2}\}, \quad (3.20)$$

where $\Delta(s)\equiv[(1-\gamma_F)^2-4\gamma_F s]^{1/2}$. Clearly $p^{-1}(s)$ remains finite for all finite values of s.

In view of the above properties one may alter the s-integration (Bromwich) contour in (3.19) so as to proceed from $-\infty$ to 0 along a line parallel to but slightly below the negative real axis, and then back to $-\infty$ on a similar line slightly above that axis. The contributions of each integral from the intervals $(-\infty\pm i0, -2\Omega_{\max}\pm i0]$ cancel, with the result

$$G(t) = -\frac{1}{\pi}\int_{-2\Omega_{\max}}^{0} ds\ e^{st}\,\mathrm{Im}\left\{p^{-1}(s+i0)\int_{-\pi}^{\pi} dq[S(q,0) - \chi(q,T_F)][s+i0+2\Omega(q)]^{-1}\right\}. \quad (3.21)$$

The function $J(q,t)$ is in principle determined by (3.6) once $G(t)$ is known. In practice, a simpler approach is to exploit the convolution form of (3.6) so as to obtain

$$J(q,t) = 2\sin^2(q/2)L_t^{-1}(q(s)/\{p(s)[s+2\Omega(q)]\}). \quad (3.22)$$

2. Asymptotic behavior

The leading asymptotic behavior of $G(t)$ for large t is easily obtained from (3.19). In this regime only the segment of the negative real s axis adjacent to the origin is of any consequence. For this segment we may replace $p(s+i0)$ by $p(0)=2\pi(1-\gamma_F^2)^{-1/2}$, $S(q,0)-\chi(q,T_F)$ by its value for $q=0$, $\Omega(q)\approx(1-\gamma_F)q^2/4$, and use the following result, which is valid for negative real s of sufficiently small magnitude,

$$\mathrm{Im}\int_{-\pi}^{\pi} dq\ \frac{1}{s+i0+(1-\gamma_F)(q^2/2)} = -\pi\left[\frac{2}{(1-\gamma_F)|s|}\right]^{1/2}. \quad (3.23)$$

Thus, the leading term for large t, of the asymptotic expansion of (3.21) is given by

$$G(t) \sim \left(\frac{1+\gamma_F}{2\pi}\right)^{1/2}[S(0,0)-\chi(0,T_F)]t^{-1/2}. \quad (3.24)$$

In particular, this function decays according to a square-root power law, as we have claimed earlier in this article. Finally, combining (3.12) and (3.24) provides an asymptotic expression for $J(q,t)$ for the regime $\Omega(q)t \geqslant 4$.

In the following section we specialize to the case $T_F=\infty$ and suppose that the system initially is in thermal equilibrium at a temperature T_I, so that $S(q,0)=\chi(q,T_I)$. We derive exact formulas for $G(t)$ and $J(q,t)$ for arbitrary values of t. The detailed analysis for arbitrary times when T_F is finite will be presented elsewhere.

IV. EXACT SOLUTION FOR $T_F=\infty$

A. Preliminaries

In Sec. II we remarked that the exact equation of motion (2.32) for $S(q,t)$, or (3.1) in terms of the dimensionless time variable, takes on a particularly simple form if the system is suddenly placed in contact with a heat bath at infinite temperature. In this case the parameter $\gamma_F=\tanh(2K_F)$ vanishes, (2.34) gives $G(t)=\Phi_1(t)$, the equation of motion (3.1) reduces to (3.3), and $G(t)$ satisfies the integral equation (3.7). The functions $P(t)$ and $Q(t)$ appearing in that equation are defined by (3.8) and (3.9), where now $\Omega(q)=\sin^2(q/2)$. The key feature is that for this choice $T_F=\infty$ without invoking any approximations, $S(q,t)$ is rigorously decoupled from higher-order spin correlation functions. In this section we derive the detailed properties of S.

We remark that with $\gamma_F=0$ several of the equations in Sec. II greatly simplify. Thus, the quantity $W_{i,i+\nu}$, given by (2.20), reduces to a constant $W_{i,i+\nu}=\alpha$, while the single-spin equation of motion (2.24) reduces to $\tilde{D}_\sigma\sigma(q)=-4\alpha\sin^2(q/2)\sigma(q)$. In particular, $\sigma(q)$ is an eigenvector of the operator $\tilde{D}_\sigma$, corresponding to the eigenvalue $-4\alpha\sin^2(q/2)$. The specific dependence of this eigenvalue on wave vector is a direct consequence of the spin-conserving dynamics of this model. That such a simple result emerges is of course due to the fact that the master equation operator $D[\sigma|\sigma']$ of (2.19), reduces to a quadratic form in the spin variables. To an extent one can rephrase this property in the language of standard many-body theory, that when $\gamma_F=0$ the quartic coupling between spins vanishes and the equation of motion can be described in terms of independent single spins. However, this analogy is not complete, since, first, the two-spin object $S(q,t)$ is not an eigenvector of the dynamics in this limit, and, second, to obtain S we must yet solve the integral equation for G so as to satisfy the global constraint (2.9). As we shall see, in the following this renders the calculation of $S(q,t)$ nontrivial. As we have remarked previously, the Glauber model is solvable without approximation for *any* value of T_F; for that model the corresponding master equation operator $D[\sigma|\sigma']$ is always quadratic in the spin variables (see Appendix B).

B. Determination of $G(t)$

In this subsection we obtain detailed results for $G(t)$ starting from the general result (3.21). For the present case of $T_F=\infty$, recalling that $\Omega(q)=\sin^2(q/2)$, we have $\Omega_{\max}=1$, as well as $\chi(q,T_F)=1$, and $S(q,0)$ is given by (2.6). We also have that (3.20) reduces to

$$p^{-1}(s)=\frac{1}{4\pi}\,[s+2+s^{1/2}(s+2)^{1/2}]. \tag{4.1}$$

It is then fairly straightforward to show that $G(t)$ may be written as

$$G(t)=(1-u)F(\theta,t), \tag{4.2}$$

where the function F is defined by

$$F(w,\tau)=\frac{1}{2\pi}\int_0^1 dx\,\frac{\exp(-2\tau x)}{x+w}\,[(1-x)/x]^{1/2} \tag{4.3}$$

for arbitrary values of the complex variable w. The quantity $\theta\equiv(1-u)^2/(4u)$ is a parametrization of the coupling strength K, which can be reexpressed in terms of the correlation length ξ as

$$\theta(K)=\begin{cases}\sinh^2[(2\xi)^{-1}] & (K>0)\\ -\cosh^2[(2\xi)^{-1}] & (K<0).\end{cases} \tag{4.4}$$

Here θ is to be evaluated using the coupling constant K corresponding to the *initial* temperature T_I. Note that $\theta>0(<-1)$, for $K>0(<0)$, respectively. To arrive at (4.3) starting from (3.21) we first performed the integration over q, noting that $(s+i0)^{1/2}=(-s)^{1/2}i$ for values of s on the segment $(-2,0)$ of the real s axis, and introduced the integration variable $x=-s/2$.

In Appendix A we discuss in detail the major properties and provide a number of useful expansions of the function F defined in (4.3). One of the most computationally effective expansions is (A21), which is of the Neumann type, with the result that (4.2) may be written as

$$G(t)=e^{-t}\left[uI_0(t)+(1+u)\sum_{k=1}^{\infty}u^kI_k(t)\right]. \tag{4.5}$$

Obviously (4.5) reduces to the correct initial value $G(0)=u$ [see (2.34)] since $I_k(0)=\delta_{k,0}$. For a given value of t, this series converges more rapidly the higher the value of the initial temperature corresponding to smaller values of the parameter u. Also note that the modified Bessel functions $I_k(t)$ decay extremely rapidly with increasing k for $k>t$. Thus, if the time t is not too large one can achieve results of high numerical accuracy by summing a relatively small number of terms in the series (4.5). However this process becomes rather lengthy for large values of t. An accurate method for calculating values of $I_k(t)$ consists of using the standard Taylor series expansion [Eq. (9.6.10) of Ref. [46]]. All terms of that series are positive, so round-off problems do not arise. However, a far more efficient procedure consists of the following. We calculate $I_0(t)$ using its Taylor series, and then use a recurrence formula [Eq. (9.6.26) of Ref. [46]] to obtain values of the ratios $r_k=I_{k+1}/I_k$ for each desired value of the argument t. To avoid crippling numerical instabilities it is necessary to invoke a backward iteration [47] method. One then has $I_{k+1}=I_0r_0r_1\cdots r_k$.

Another numerically effective and physically insightful approach for calculating $G(t)$ for $t>4$ consists of employing (A23) for $K>0$, and (A24) for $K<0$. The details of this approach are given in the following paragraphs.

1. Ferromagnetic spin coupling

We consider first the case of ferromagnetic spin interactions ($K>0$). Using (4.2) and (A23) we have for $t \gg 4$

$$G(t) \sim u^{1/2} \exp(t/t_w) \operatorname{erfc}[(t/t_w)^{1/2}], \tag{4.6}$$

where $t_w \equiv (2\theta)^{-1}$. Numerical comparisons between the approximate result (4.6) and the exact Neumann expansion (4.5) shows that the former provides an *excellent* approximation to $G(t)$ for times satisfying $t>4$, although the accuracy depends somewhat on the initial temperature T_I. Note that the $t=0$ value of (4.6) is $\sqrt{u}$, whereas the exact result is given by $G(0)=u$. Thus, the lower the value of the initial temperature, the closer are the initial values for the two cases, and the better that (4.6) approximates (4.2), even for $t<4$.

The formula (4.6) exhibits an important scaling property, namely, that the time evolution of the nonequilibrium nearest-neighbor correlation function is expressible solely in terms of the dimensionless variable t/t_w. The physical origin of the characteristic time t_w is as follows. In the initial state, where the system is in equilibrium at a temperature T_I, the ferromagnetic spin coupling gives rise to ordered domains with a characteristic size of the order of the initial correlation length $\xi^{-1}=\ln(\coth K)$ governing the exponential decay of the equilibrium correlation function (2.3). For low initial temperatures $\xi \approx \exp(2K)$. Upon suddenly raising the temperature to $T_F=\infty$, the domains begin to decompose as a result of spin exchanges, which, we note, initially occur solely at the *boundaries* between the domains of oppositely aligned spins. Because of the form of the dynamics for $K_F=0$, the spin-exchange probability is independent of the local spin configuration [see (2.20)]. Hence if the nearest-neighbor pair of spins at the domain boundaries have flipped, there is equal probability of either recovering their previous configuration, or for continuing spin exchanges to gradually propagate into the interior of the two original domains. This can be pictured in terms of the domain boundaries performing independent random walks [48]. One therefore expects that the time to randomize a domain of size ξ by spin exchanges will be on order of $\alpha^{-1}\xi^2 \approx (\alpha\theta)^{-1}$, and the latter is consistent with our definition of the (dimensionless) time t_w. In short, the scaling property explicit in (4.6) is an expression of the fact that the constraint of a conserved order parameter ensures that the time evolution of the ferromagnetic spin correlations proceeds by the random walk of domain boundaries as the mechanism for the demise of the domains. We can then see from (3.14) that for long times the evolution of $S(q,t)$ will also exhibit this scaling property. In the absence of the requirement of a conserved order parameter, the domains would be disrupted by spontaneous spin flips within the interior of the domain, and this scaling property would not hold for such systems. Similar statements can be expected to apply for the two- and three-dimensional variants of this model.

The limiting form of (4.6) for times such that $t \gg t_w$ is given by

$$G(t) \sim \frac{u}{1-u} (\pi t/2)^{-1/2}, \tag{4.7}$$

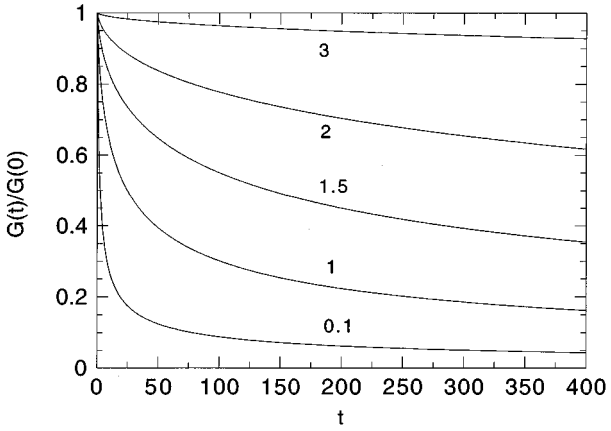

FIG. 2. Time dependence of the nonequilibrium nearest-neighbor correlation function $G(t)/G(0)$ for ferromagnetic spin coupling, as given by (4.2). Each curve is labeled by the value of K used in the calculation.

and we obtain the expected $t^{-1/2}$ time dependence. Equation (4.7) is in accord with (3.24) in this limit. Note that the value of t_w grows extremely rapidly as the initial temperature is reduced. For example, the values of t_w for $K=0.1$, 1.0, 1.5, 2.0, and 3.0 are 0.24, 26.8, 200, 1 488, and 81 376, respectively. Thus, although the scaling form of (4.6) is already in effect for $t \gg 4$, the power-law form (4.7) becomes operative for the low-temperature regime only for *extremely* late times.

In Fig. 2 we show the time dependence of $G(t)/G(0)$ computed from (4.5) for ferromagnetic ($K>0$) spin interactions. Each curve is labeled by the value of K employed in the calculation. The curves we would obtain using the scaling formula (4.6) are indistinguishable in the figure from those shown. As noted in the preceding paragraph, for $K=2,3$ the values of t_w are so large as to be off the scale shown in Fig. 2. The existence of such long waiting times for the ferromagnetic system, initially prepared at low temperatures, despite the fact that the system has been placed in contact with a heat bath at temperature $T_F=\infty$, is a direct consequence of the spin-exchange dynamics of the model.

2. Antiferromagnetic spin coupling

We now provide the analog of (4.6) for the case of antiferromagnetic interactions ($K<0$) so that θ, given by (4.4), satisfies the inequality $\theta<-1$. Using (4.2) as well as the asymptotic formula (A24), we obtain as the leading behavior in the regime $t>4$

$$G(t) \sim \frac{-|u|}{1+|u|} (\pi t/2)^{-1/2}. \tag{4.8}$$

Superficially, the asymptotic formulas (4.7) and (4.8) appear to agree. Note however, that (4.7) applies only for $t \gg t_w$. The waiting time t_w is very large, for the same value of T_I, compared to the time $t \approx 4$ where (4.8) applies. That is, in the antiferromagnetic case there is no significant waiting time for the domains to randomize, and one obtains the power-law form almost immediately, namely, for $t>4$. These properties are evident in Fig. 3 where we show the time dependence of $G(t)/G(0)$ for $K<0$. The solid curves correspond to the ex-

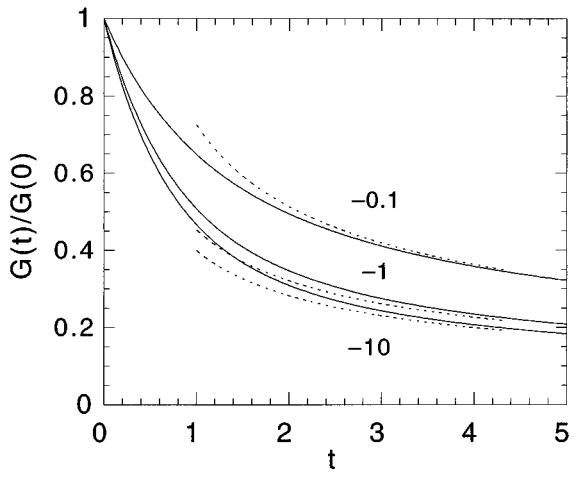

FIG. 3. Time dependence of $G(t)/G(0)$ for antiferromagnetic spin coupling as given by the exact result (4.2), (solid curves), as well as the asymptotic formula (4.8), (dashed curves), for the labeled values of K. The asymptotic regime commences for $t\approx 4$.

act values of $G(t)/G(0)$, while the dashed curves are obtained using the asymptotic expression (4.8). Note that the power-law form is well obeyed for $t>4$. We note that in contrast to the ferromagnetic case, $G(t)$ decays *faster* the lower the initial temperature. This is to be expected since the more ordered the initial antiferromagnetic state, the more rapidly the structure is disrupted by the near-neighbor spin-exchange process.

C. Determination of $S(q,t)$

The remaining task to be performed so as to determine $S(q,t)$ is to establish the form of the function $J(q,t)$ given by (3.6). Once this is achieved we have [see (3.5)]

$$S(q,t)=1+[\chi(q,T_I)-1]\exp[-2t\,\sin^2(q/2)]-J(q,t). \tag{4.9}$$

Probably the simplest way to proceed is to substitute in (3.6) for the function G using (4.2) and (4.3) and to interchange the order of the integrations. The calculation is fairly straightforward upon using the identity

$$\int_0^1 dx\left(\frac{1-x}{x}\right)^{1/2}\frac{1}{x+w}=\pi\left[\left(\frac{1+w}{w}\right)^{1/2}-1\right], \tag{4.10}$$

which applies for arbitrary values of the complex variable w. For the special case that w approaches the interval $(-1,0)$ of the real axis, (4.10) can be used to yield the relation

$$\mathrm{P}\int_0^1 dx\left(\frac{1-x}{x}\right)^{1/2}\frac{1}{x+w}=-\pi, \tag{4.11}$$

where P denotes principal value. With the aid of these identities and exploiting a partial fraction decomposition of the rational form $1/[(x+\theta)(x-\sin^2 q/2)]$, one finds

$$J(q,t)=\frac{\sin^2 q/2}{\theta+\sin^2 q/2}\left\{(1-u)\{F(\theta,t)-\mathrm{Re}\,F[-\sin^2(q/2)\pm i0,t]\}-\frac{1}{2}(1+u)e^{-2t\,\sin^2 q/2}\right\}. \tag{4.12}$$

As is evident from (3.6), $J(q,0)$ must vanish. Using (A4) and (A22) one finds $F(\theta,0)=u/(1-u)$ and $F[-\sin^2(q/2),0]=-1/2$, so that this requirement is satisfied.

Combining (4.12) with (4.9) gives $S(q,t)$. These formulas can be simplified greatly in the case $q=\pi$. Indeed, we have special interest in the time dependence of $S(\pi,t)$ for antiferromagnetic interactions. For this special value of q, starting from (A16), one can show that

$$S(\pi,t)=1+[\chi(\pi,T_I)-1]e^{2\theta t}-(1-u)t\frac{d}{dt}[e^{-t}I_0(t)]+O((1+u)^2). \tag{4.13}$$

This formula is useful for all t and for very small values of T_I, where we have $\theta\approx -1$ and $u\approx -1$ for antiferromagnetic interactions. We shall use it in our discussion in the following paragraph. Note that the term containing the factor $t(d/dt)[e^{-t}I_0(t)]$ may be approximated as $(2\pi t)^{-1/2}$ for $t\geq 2$, explicitly showing once again the slow power-law decay that applies after the initial Bragg peak has decayed sufficiently.

We now summarize our major results for $S(q,t)$. In Fig. 4(a) we show values of $S(q,t)$ as a function of wave vector for the antiferromagnetic with initial coupling $K_I=-3$ in the immediate vicinity of the BZ boundary for the dimensionless times 0, 0.25, 0.5, 1.0. The numerical values of $S(q,t)$ were obtained using (4.9), (4.12), and the Neumann expansions (A21) and (A22). The calculated values for the special case $q=\pi$ are in excellent quantitative agreement with (4.13), which is to be expected since the coefficient $(1+u)^2$ of the neglected correction term in that equation is 2.4×10^{-5}. The rapid decrease of the initial Bragg peak with time is noteworthy. The qualitative discussion in Sec. III C 1 leads to the conclusion that in the first stage this decrease should proceed in an exponential fashion, with a characteristic time that is essentially independent of wave vector. In particular, the shape of the decaying Bragg peak should be preserved and a semilog plot of $S(q,t)/\chi(q,T_I)$ should exhibit a linear dependence on t. This predicted behavior is confirmed by the data shown in Fig. 4(b) for early times, approximately $t\leq 1.5$. Subsequently the curves for different wave vectors fan out and the decay proceeds at a much slower rate. In this time regime the further decay of the Bragg peak should proceed as $t^{-1/2}$. This feature is in fact supported by our computed data.

In Fig. 5 we provide data for $S(q,t)$ for all wave vectors in the BZ, again for $K_I=-3$. The approximate behavior in the regime where $t\,\sin^2(q/2)$ is small compared to unity has been discussed in Sec. III C 3, and the results are summarized by (3.16). For $|q|\leq 0.8\pi$ the first term in (3.16), the initial susceptibility, is negligibly small, and we can expect that the q dependence will be proportional to $\sin^2(q/2)$. The amplitude of this term is given by $2[t-\int_0^t dt'\ G(t')]$, which is dominated by linear growth. The data shown in Fig. 5 are consistent with these predictions. As time progresses and

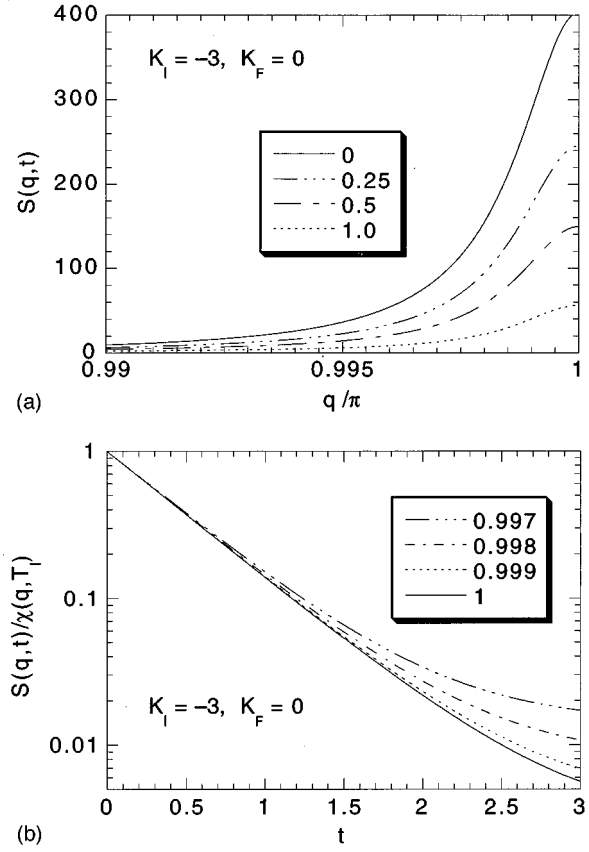

FIG. 4. (a) Wave-vector dependence of the nonequilibrium structure factor $S(q,t)$, (4.9), in the vicinity of the Bragg peak, for the times listed in the legend, for spins with antiferromagnetic spin coupling initially in thermal equilibrium (initial coupling constant $K_I=-3$) that suddenly (at $t=0$) are brought in contact with a heat reservoir of infinite temperature ($K_F=0$). (b) Semilog plot of the decay of $S(q,t)/\chi(q,T_I)$, for the system described in (a), in the region of the Bragg peak, as a function of t for values of q/π listed in the legend. The initial decay of S is exponential, but the time period over which this behavior persists decreases with decreasing q. In the regime of exponential decay, the shape of $S(q,t)$ remains approximately invariant.

$t\sin^2(q/2)$ is no longer small compared to unity, in fact when $\Omega(q)t\geq 4$, we may use (3.14). Except in the immediate vicinity of the BZ boundary we may neglect the first term of that equation. For the present case of $T_F=\infty$ we have $\gamma_F=0$ and $\chi(q,T_F)=1$ for all q. Thus for $\Omega(q)t\geq 4$, we have $S(q,t)\approx 1-G(t)$. In particular S is independent of q and very slowly (like $t^{-1/2}$ for large t), *decreases* towards unity, because $G(t)<0$ for antiferromagnetic interactions. This predicted plateaulike behavior is confirmed by the data shown for $t\geq 4$. As expected, even for the time $t=400$, for $|q|\approx 0$ the structure factor remains ''frozen'' at its initial value, since for a small value of q, one reaches the regime $\Omega(q)t\geq 4$ only when $t\geq 16q^{-2}$. This behavior is due to the combined effects of the vanishing of $\Omega(q)$ like q^2 for small q and the requirement of the sum rule (2.9).

In Fig. 6 we display our results for ferromagnetic interactions for initial coupling $K_I=3$. The persistence of the Bragg peak for $|q|\approx 0$ is very dramatic and, as discussed in Sec. III C 1, it follows from the fact that although the decay is exponential, the corresponding time constant is given by

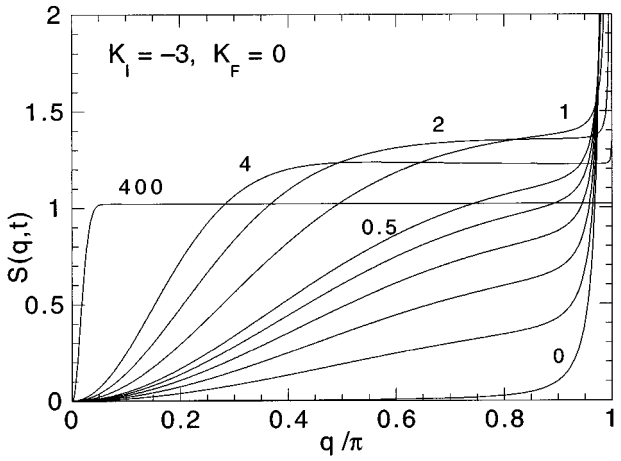

FIG. 5. Wave-vector dependence of the nonequilibrium structure factor $S(q,t)$ for the system described in Fig. 4 but for the entire Brillouin zone. The behavior of S can be described as though the spins for the larger values of q rapidly equilibrate to the final structure factor at $T_F=\infty$, for which S is independent of q, but with a slowly decaying time-dependent amplitude. This slow decay is a direct consequence of the conserved spin dynamics and the sum rule (2.9). Similarly the behavior for $q\approx 0$ can be described as though the spins remain ''frozen'' at their initial structure factor because of the conserved spin dynamics.

$\tau_q\sim 2q^{-2}$, which diverges for $q\to 0$. For $|q|\geq 0.2\pi$ the displayed data shows plateaulike behavior $S(q,t)\approx 1-G(t)$ since $\Omega(q)t\geq 4$ for the times considered. An increase towards unity is to be expected since G is positive for all t. However, this rate of increase towards unity is extremely small, because of the corresponding slow decay of the Bragg peak along with the constraint of the sum rule (2.9). As discussed in Sec. IV B 1, the function $G(t)$ is given by (4.6).

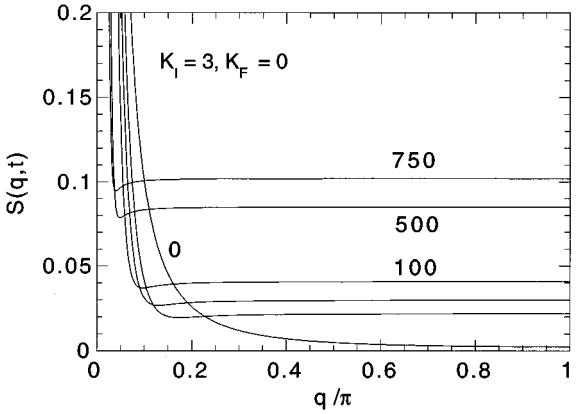

FIG. 6. Wave-vector dependence of $S(q,t)$, for the dimensionless times 0, 25, 50, 100, 500, 750, for spins with ferromagnetic spin coupling initially in thermal equilibrium (initial coupling constant $K_I=3$) that are suddenly brought in contact with a heat reservoir of infinite temperature ($K_F=0$). The main features of the behavior are qualitatively similar to the case of antiferromagnetic coupling (Fig. 5). For larger values of q the spins rapidly equilibrate to the final temperature structure factor, but with an amplitude which decays in an extremely slow manner, whereas for $q\approx 0$ it is as though the spins are frozen at their initial structure factor.

The asymptotic form (4.7) (square-root decrease) becomes operational only for $t \gg t_w$, where $t_w = 81\,376$.

We remind the reader that the major qualitative characteristics of S were already established in Sec. III C, without the need for evaluating the functions $G(t)$ and $J(q,t)$, which are given by (3.21) and (3.22). Of course, one could not avoid evaluating the latter equations in order to establish the detailed quantitative behavior of S.

V. SUMMARY

In this article we have investigated the time evolution of the nonequilibrium structure factor $S(q,t)$ for a system of spins subject to Kawasaki spin-exchange dynamics, which conserves the total spin. We have considered the case where the system is subject to a sudden temperature increase, from an initial temperature T_I to a final temperature T_F. The existence of a conserved variable greatly complicates the treatment, as compared to the analogous issues for the Glauber dynamics, which does not possess a conserved mode. Whereas an exact expression for $S(q,t)$ can be derived for the latter model in one dimension whatever the values of T_I and T_F, for the case of the spin-conserving dynamics, an approximation-free treatment can be given only if T_F is infinitely large. Except for that case, the equation of motion satisfied by $S(q,t)$ involves a nonterminating hierarchy of equations of motion for higher-order spin correlation functions.

We have found the exact form of $S(q,t)$ for the case $T_F = \infty$ in Sec. IV, which, to the best of our knowledge, is the first instance of an exact solution for $S(q,t)$ in which the total spin is conserved. In order to gain insight for the intractable case of finite T_F, we have invoked an approximation procedure that truncates the hierarchy of equations of motion at the level of two-spin nonequilibrium correlation functions. This truncation procedure was introduced in Sec. III B and consists of replacing the four-spin nonequilibrium correlation function $\langle \sigma_{-1}\sigma_0\sigma_1\sigma_n \rangle_t$ by the two-spin quantity $\langle \sigma_0\sigma_n \rangle_t$, leading to a solvable equation of motion for $S(q,t)$. Although invoking uncontrolled approximations in nonequilibrium problems can have profound effects, we believe our approximation is physically quite reasonable, and arguments in support of this approximation were presented in that section. The approximate equation of motion resulting from our truncation procedure was shown to preserve the following two crucial features of the exact solution: (i) $S(q,t)$ evolves to the correct long-time value $\chi(q,T_F)$ the final-state equilibrium structure factor, and (ii) $S(q,t)$ satisfies the exact sum rule (2.9).

The formal solution of the resulting approximate equation of motion for $S(q,t)$ was obtained in Sec. III D for arbitrary T_F. The details were worked out in Sec. IV only for the special case of infinite T_F. We plan to work out the *quantitative* details for the case of finite T_F in a future publication. However, even in the absence of such a treatment, it was already possible in Sec. III C to outline the qualitative features and major trends in the behavior of $S(q,t)$. We briefly summarize the major conclusions.

First, as discussed in Sec. III C 1, in the initial stage the decay of a Bragg peak proceeds exponentially for either ferromagnetic or antiferromagnetic spin interactions. The length of this stage increases without bound as T_I is decreased towards zero. In the case of antiferromagnetic interactions one can, additionally, speak of the Bragg peak maintaining its shape as a function of wave vector, during this initial stage of exponential decay. This is a consequence of the fact that the lifetime τ_q is in essence independent of wave vector in the immediate vicinity of the BZ boundary. For the case of ferromagnetic interactions, however, although the Bragg peak for $q \approx 0$ also initially decays exponentially with time, in the process the shape of the peak is not retained. This is because τ_q is strongly dependent on q; diverging as q^{-2} for small q.

Second, as discussed in Sec. III C 2, as time progresses $[\Omega(q)t \geq 4]$, $S(q,t)$ demonstrates quasiequilibrated behavior. Specifically, apart from a time-dependent amplitude, $S(q,t)$ shows the same *wave-vector dependence* as its equilibrium form at the final temperature, i.e., $S(q,t)/\chi(q,T_F) \approx A(t)$, independent of wave vector but dependent on time. The function $A(t)$ approaches unity at long times with a $t^{-1/2}$ correction term. This description is applicable for both ferromagnetic and antiferromagnetic interactions, except that in the latter case for wave vectors in the immediate vicinity of the BZ boundary the presence of the Bragg peak complicates the story. [In particular, if the initial temperature is sufficiently small, the behavior of $S(q,t)$ continues to be dominated by the exponentially decaying Bragg peak long after the behavior $S(q,t)/\chi(q,T_F) \approx A(t)$ has set in for smaller values of $|q|$.] As time progresses, the range of wave vectors for which this description is valid gradually spreads towards the interior of the BZ. By contrast, for wave vectors in the vicinity of the center of the BZ, the behavior of S over long periods of time is as though the spins are frozen at their initial temperature, and thus for these wave vectors $S(q,t) \approx \chi(q,T_I)$. The physical origin of this very slow evolution for small wave vectors is due to the fact that achieving a major change in S for small q requires a long-range spatial reorientation of the spins, but such a process is perforce very slow since a given spin will flip only if one of its two nearest-neighbors is at that moment oriented oppositely. This process can be visualized in terms of random walks of domain boundaries, which feature spatial progression on a lattice proportional to the square root of the time interval.

Thus far our remarks have been restricted to our explicit results for the one-dimensional spin-exchange model. We believe, however, that these results can offer qualitative insights into the disordering behavior of higher-dimensional systems, e.g., the disordering of adsorbed monolayers upon a sudden increase in temperature at constant coverage [49]. This is because the characteristic features of $S(q,t)$ for disordering are shaped first and foremost by the requirement of a dynamics that features a conserved mode. The dimensionality of the system is largely a secondary issue, with the exception that at long times we expect the dimensionality to be manifested in a $t^{-d/2}$ power-law decay, the signature of diffusive motion in d dimensions. Of course, the *equilibrium* properties of a higher-dimensional lattice of Ising spins are profoundly different from those for its one-dimensional counterpart. Nevertheless we believe that the impact of the constraint of the conserved total spin on the time evolution is of prime importance as compared to the spatial dimension of the system.

It would be of great benefit if this qualitative description of the dynamics could be put to experimental test. A two-dimensional spin-exchange model should be relevant in describing diffusive processes of atoms on surfaces where both adsorption and desorption are absent. We are aware of only one experiment where the shape-preserving exponential decay of the Bragg peak has been observed. This is the case of the disordering of the Si(100)$-(2\times n)$ ordered-defect state [19]. We have also found some tentative evidence for this type of behavior upon analyzing some Monte Carlo data (Fig. 3 of Ref. [17]) for the two-dimensional Ising model with nearest-neighbor repulsion having the $c(2\times 2)$ phase as its ground state. We also urge that our prediction for sufficiently long times, namely, that $S(q,t)/\chi(q,T_F)\approx A(t)$ independent of wave vector, be subject to experimental test. If confirmed it would be of great interest to test whether the long-term time dependence of $A(t)$ is that of t^{-1}, appropriate to two dimensions.

The disordering process resulting from an abrupt increase in the temperature involves issues in nonequilibrium statistical mechanics of great difficulty yet great interest. To provide an approximation-free treatment based on spin-exchange processes we had to restrict our attention to a one-dimensional system and whose final temperature is infinitely large. Nevertheless, the approximate treatment developed in Sec. III has provided us with clear expectations for higher-dimensional systems and where T_F is finite, even though an approximation-free treatment is out of reach. One can anticipate that the availability of accurate experimental data for the nonequilibrium structure factor would spawn significant theoretical progress in understanding these issues.

ACKNOWLEDGMENTS

We thank M. C. Tringides for initially raising some of the experimental issues that led to the present investigation and several subsequent discussions. We also thank C. M. Bender for a helpful discussion regarding the asymptotic behavior of the dispersion integral treated in Appendix A. Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under contract W-7405-Eng-82.

APPENDIX A: THE INTEGRAL $F(w,\tau)$

1. Preliminaries

In this Appendix we establish the major properties and useful expansions of the function defined for arbitrary complex w by

$$F(w,\tau)=\frac{1}{2\pi}\int_0^1 dx\left(\frac{1-x}{x}\right)^{1/2}\frac{\exp(-2\tau x)}{x+w}\quad(\mathrm{Re}\tau>0). \tag{A1}$$

This function first appears in Sec. IV, in (4.2), where $w=\theta$, a quantity defined by (4.4), and where $\tau=t$, the dimensionless time variable. We recall that $\theta>0$ for ferromagnetic coupling, whereas $\theta<-1$ for antiferromagnetic coupling. For these (real) values of the argument w the function F is real. Additionally, in (4.12) we require ReF for values of w which are approaching the real interval $[-1,0]$, specifically for $w=-\sin^2(q/2)\pm i0$, where q lies in the BZ. This integral can also be shown to arise in the treatment of the one-dimensional Glauber model for arbitrary initial and final temperatures [50], and in Ref. [51] [see their Eq. (55)] in a study of the kinetics of a sequence of first-order chemical reactions. We have therefore chosen to provide here a list of useful expansions of this function.

The dispersion integral (A1) defines a function that is analytic in the complex w plane except for a cut in the interval $[-1,0]$, and which is also analytic in the finite part of the complex τ plane. If w is any real number in the interval $[-1,0]$ we have

$$\mathrm{Re}F(w\pm i0,\tau)=\frac{1}{2\pi}\,\mathrm{P}\int_0^1 dx\left(\frac{1-x}{x}\right)^{1/2}\frac{\exp(-2\tau x)}{x+w}, \tag{A2}$$

$$\mathrm{Im}F(w\pm i0,\tau)=\mp\frac{1}{2}\left(\frac{1+w}{-w}\right)^{1/2}e^{2\tau w}. \tag{A2'}$$

In writing (A2) and (A2′) we have used the standard identity

$$\frac{1}{x+w\pm i\varepsilon}=\mathrm{P}\,\frac{1}{x+w}\mp i\pi\delta(x+w),$$

where ε is a real, positive infinitesimal, and P denotes principal value.

For a few special cases one can evaluate (A1) in closed form. These include $w=-1$ for arbitrary τ, as well as $\tau=0$ for arbitrary w. The results are

$$F(-1,\tau)=-\frac{1}{2}\,e^{-\tau}I_0(\tau), \tag{A3}$$

where I_0 denotes a modified Bessel function, and

$$F(w,0)=\frac{1}{2}\left[\left(\frac{1+w}{w}\right)^{1/2}-1\right]. \tag{A4}$$

Note that (A4) exhibits in an explicit manner the branch points of F at $w=0,-1$. For general values of w and τ there is no alternative but to develop assorted expansions.

One can readily arrive at alternate integral representations of F that are not only of intrinsic interest but will greatly aid us later in developing expansions for various regimes. Suppose first that Re$w>0$. We may then substitute in (A1) the integral representation

$$(x+w)^{-1}=\int_0^\infty ds\,\exp[-s(x+w)]\quad(\mathrm{Re}w>0). \tag{A5}$$

Interchanging the order of integrations and using the identity [52]

$$\int_0^1 dx\,\exp(-zx)\left(\frac{1-x}{x}\right)^{1/2}=\left(\frac{\pi}{2}\right)e^{-z/2}[I_0(z/2)+I_1(z/2)], \tag{A6}$$

we arrive at the formula

$$F(w,\tau)=e^{2\tau w}\left\{F(w,0)-\frac{1}{2}\int_0^\tau dx\ e^{-(2w+1)x}\times[I_0(x)+I_1(x)]\right\}. \quad \text{(A7)}$$

Recall that $F(w,0)$ is given by (A4). We emphasize that the result (A7), although derived subject to the restriction $\mathrm{Re}w>0$ so as to write (A5), can be analytically continued throughout the w plane.

A useful check of (A7) is provided by considering the special case $w=-1$. Using Eq. (11.3.14) of Ref. [46], the indefinite integral can now be evaluated in closed form, with the result being

$$\int_0^\tau dx\ e^x[I_0(x)+I_1(x)]=e^\tau I_0(\tau)-1. \quad \text{(A8)}$$

The resulting value of $F(-1,\tau)$ obtained from (A7) agrees with (A3).

A useful variant of (A7) can be obtained by rewriting the integral from 0 to τ as the integral from 0 to ∞ minus the integral from τ to ∞. This leads to the result

$$F(w,\tau)=\frac{1}{2}\,e^{2\tau w}\int_\tau^\infty dx\ e^{-(2w+1)x}[I_0(x)+I_1(x)] \quad (\mathrm{Re}w>0). \quad \text{(A9)}$$

We emphasize that (A9) is applicable only for the right-half w plane, as is evident from the fact that the integral diverges if $\mathrm{Re}w<0$ since the functions $I_{0,1}$ grow as $e^x(2\pi x)^{-1/2}$ for large positive x. Each of (A7) and (A9) will be employed in Sec. A 5 as the starting point for developing useful approximate expressions for F for large values of τ.

We now provide two integral representations of F which involve closed integration contours. The advantage provided by a closed contour is that one can employ the Cauchy residue theorem and other standard results of analytic function theory.

The first of these representations is obtained by starting from (A1) and replacing the given integration contour by the double hairpin contour which encloses the closed interval $[0,1]$ of the real x axis and which is traversed in the *negative* (clockwise) sense. It is assumed that $-w$ lies outside the hairpin, i.e., w does not lie on the interval $(-1,0)$ of the real axis. On the upper line segment of the hairpin we have $\arg x=\arg(1-x)=0$, whereas on the lower line segment $\arg x=0$, $\arg(1-x)=-\pi$. Hence the contribution from the lower line segment is identical to that from the upper segment. It therefore follows that

$$F(w,\tau)=\frac{1}{4\pi}\oint dx\,\frac{e^{-2\tau x}}{x+w}\left(\frac{1-x}{x}\right)^{1/2}. \quad \text{(A10)}$$

The second closed contour representation is obtained from (A10) by defining a new variable z according to the relation $x=-(z-1)^2/(4z)$. Under this mapping the hairpin contour in (A10) corresponds to traversing the unit circle in the z plane and in the *positive* (counterclockwise) sense. It is then straightforward to show that (A10) may be rewritten as

$$F(w,\tau)=\frac{ie^{-\tau}}{4\pi}\oint dz\,\frac{(z+1)^2}{z[z^2-2(2w+1)z+1]}\times\exp\frac{\tau}{2}\left(z+\frac{1}{z}\right). \quad \text{(A11)}$$

Note that for the special case $w=-1$ the last formula reduces to

$$F(-1,\tau)=\frac{ie^{-\tau}}{4\pi}\oint dz\,\frac{1}{z}\exp\frac{\tau}{2}\left(z+\frac{1}{z}\right). \quad \text{(A12)}$$

This integral can immediately be evaluated by noting that the exponential term in the integrand is the generating function for modified Bessel functions of integral order [Eq. (9.6.33) of Ref. [46]]

$$\exp\frac{\tau}{2}\left(z+\frac{1}{z}\right)=\sum_{n=-\infty}^{\infty}z^nI_n(\tau). \quad \text{(A13)}$$

Interchanging the sum and integration in (A12), noting that the only nonzero contribution is from the $n=0$ term, and using the Cauchy residue theorem, we find that (A12) agrees with (A3). This procedure of utilizing (A13) will be generalized in Sec. A 4 to provide an expansion of $F(w,\tau)$ for arbitrary (in general, complex) values of w, as an infinite series in the I_n.

2. Taylor expansion in τ

The function F can be expanded as a Taylor series in powers of τ, (A16), which has an infinite radius of convergence. To obtain this result we replace $e^{-2\tau x}$ in (A1) by its power series expansion and integrate term by term. One readily finds

$$F(w,\tau)=\frac{1}{4w}\sum_{n=0}^{\infty}\frac{(1/2)_n}{(2)_n n!}\,{}_2F_1\left(1,n+\frac{1}{2};n+2;-1/w\right)\times(-2\tau)^n, \quad \text{(A14)}$$

upon using the integral representation of the hypergeometric function ${}_2F_1(a,b;c;z)$ [Eq. (15.3.1) of Ref. [46]]. Here $(a)_0=1$, $(a)_n=a(a+1)\cdots(a+n-1)$ is the Pochammer symbol. It will be recalled that the hypergeometric function has branch points at $+1$ and ∞, and possesses a power series expansion in z with unit radius of convergence [Eq. (15.1.1) of Ref. [46]]. Noting these analytic properties of the hypergeometric function, the individual terms of the expansion (A14) are seen to exhibit the branch points ($w=0,-1$) of F. For actual numerical calculations, the power series representation of F could be used to evaluate the expansion coefficients in (A14) as long as $|w|>1$. However, rather than be limited by this restriction on $|w|$, it is actually preferable to derive, starting from (A14), an alternate expansion of F which can be used for all w. We employ the identity

$$ {}_2F_1\left(1,n+\frac{1}{2};n+2;-1/w\right)=\frac{2w(2)_n(-w)^n}{(1/2)_n}\left(\frac{1+w}{w}\right)^{1/2} -2w(n+1){}_2F_1 \times\left(1,-n;\frac{1}{2};1+w\right) \quad \text{(A15)} $$

which can be derived using Eqs. (15.3.6) and (15.3.7) of Ref. [46], so that (A14) may be rewritten in the more convenient form

$$ F(w,\tau)=\frac{1}{2}\left(\frac{1+w}{w}\right)^{1/2}e^{2w\tau}-\frac{1}{2}\sum_{n=0}^{\infty}\frac{\left(\frac{1}{2}\right)_n}{(n!)^2}{}_2F_1 \times\left(1,-n;\frac{1}{2};1+w\right)(-2\tau)^n. \quad \text{(A16)} $$

In contrast to (A14), which is useful for computational purposes only for $|w|>1$, (A16) can be used for all w since the power series expansion of ${}_2F_1(1,-n;1/2;1+w)$ terminates after $n+1$ terms, i.e., it is a polynomial of degree $n+1$ in the variable w. Additionally, the first term on the right-hand side of (A16) explicitly embodies all of the multivalued properties of F as a function of the variable w. In particular, the branch points of F at $w=-1$, 0 are manifestly explicit in that term. Furthermore, the series in (A16) is single valued as one crosses the branch line $[-1,0]$ of F in the variable w. In short, (A16) is especially useful for small values of τ, but it may be used for any value of w. Note that (A16) correctly reduces to (A4) upon substituting $\tau=0$. It should also be noted that (A16) reduces to (A3) upon substituting $w=-1$ since the right-hand side of (A16) may be identified with $-1/2{}_1F_1(1/2;1;-2\tau)=-1/2e^{-\tau}I_0(\tau)$. Here ${}_1F_1$ denotes the confluent hypergeometric function and we have used Eq. (13.6.3) of Ref. [46].

3. Laurent expansion in w

Our second major result is the Laurent expansion of F, in powers of w^{-1}, which in view of the location of the branch line of F, converges for the annular region $|w|>1$,

$$ F(w,\tau)=\frac{1}{4w}\sum_{n=0}^{\infty}\frac{(1/2)_n}{(2)_n}{}_1F_1\left(n+\frac{1}{2};n+2;-2\tau\right)(-1/w)^n. \quad \text{(A17)} $$

To obtain (A17) note that if $|w|>1$ the factor $(x+w)^{-1}$ in (A1) may be expanded as a geometric series in powers of x/w, which converges for all x in the integration interval. Substituting that expansion, integrating term by term, and using the integral representation of the confluent hypergeometric function, ${}_1F_1(a;b;z)$, Eq. (13.2.1) of Ref. [46] gives (A17). The power series expansion of ${}_1F_1$ converges for all finite z [see Eq. (13.1.2) of Ref. [46]].

4. Neumann expansions

We now derive an expansion of $F(w,\tau)$ as an infinite series of modified Bessel functions in the variable τ. This expansion is analogous to a Neumann [53] expansion of a function, as an infinite series of ordinary Bessel functions. It is of great computational usefulness especially for small and moderate real values of τ.

Our starting point is the integral representation (A11), and w is any complex number subject to the restriction that it not lie in the interval $[-1,0]$ of the real axis. We note that the polynomial $z^2-2(2w+1)z+1$ has two roots, $2w+1\pm2\sqrt{w(w+1)}$, one of which, to be denoted by ν, necessarily lies within the interior of the unit circle, and the second is given by $1/\nu$ and it lies exterior to the unit circle. (Subsequently we shall also allow w to approach the interval $[-1,0]$, so that both of ν and $1/\nu$ approach the unit circle, but that will be a special limiting case of the general treatment we now give.) The integration contour in (A11) is the unit circle of the z plane, and it therefore encloses the simple pole of the integrand at $z=\nu$ as well as the essential singularity at the origin. The integrand is otherwise analytic within the unit circle. We may thus alter the integration contour so as to consist of two arbitrary nonintersecting closed contours lying within the unit circle, the first enclosing the simple pole and the second enclosing the origin. The contribution of the first (pole) integral to $F(w,\tau)$, to be denoted by F_p follows immediately from the residue theorem, and it is given by

$$ F_p=\frac{1}{2}e^{-\tau}\frac{1+\nu}{1-\nu}\exp\left[\frac{\tau}{2}(\nu+1/\nu)\right]. \quad \text{(A18)} $$

To evaluate the contribution, to be denoted by F_{es}, of the second (essential singularity) integral to $F(w,\tau)$, we replace the exponential factor of the integrand by (A13) and integrate term by term, and use the fact that

$$ \frac{1}{2\pi i}\oint dz\,\frac{1}{z^n(\nu-z)}=\frac{1}{\nu^n} \quad \text{(A19)} $$

if n is a positive integer, whereas this integral equals zero if n is zero or a negative integer. [Note that the contour in (A19) encloses the origin but excludes the point ν.] One finds that

$$ F_{\rm es}(w,\tau)=\frac{1}{2}e^{-\tau}\left[-I_0(\tau)+\frac{1+\nu}{1-\nu}\sum_{n=1}^{\infty}(\nu^n-\nu^{-n})I_n(\tau)\right]. \quad \text{(A20)} $$

Finally, adding the contribution of (A18) after using (A13), one obtains the result

$$ F(w,\tau)=\frac{e^{-\tau}}{1-\nu}\left[\nu I_0(\tau)+(1+\nu)\sum_{n=1}^{\infty}\nu^n I_n(\tau)\right]. \quad \text{(A21)} $$

Note that if w is real we have $\nu=2w+1\mp2\sqrt{w(w+1)}$, where the upper (lower) sign is chosen if $w>0$ ($w<-1$).

Finally, we can use (A18) and (A20) to obtain ReF and ImF for values of w which are situated an infinitesimal distance above or below the segment $(-1,0)$ of the real axis. We write $w=-\sin^2(q/2)+i\varepsilon$, where ε is a vanishingly small real number (either positive or negative) and $0\leqslant q\leqslant\pi$. One then easily finds that $\nu\to e^{\mp iq}$, where the upper (lower) sign applies if ε is positive (negative). Using (A18) and (A20) we find that the quantities F_p and F_{es} turn out to be pure imaginary and pure real, respectively. We thus have

$$\mathrm{Re}F[-\sin^2(q/2)\pm i0,\tau]=-e^{-\tau}\left[\frac{1}{2}I_0(\tau)+\cot(q/2)\sum_{n=1}^{\infty}I_n(\tau)\sin nq\right]. \tag{A22}$$

The value of $\mathrm{Im}F[-\sin^2(q/2)\pm i0,\tau]$ in essence has already been given by (A2′).

5. Large-τ expansions

As one increases τ and enters the regime $\tau\gg 1$, the expansions (A21) and (A22) gradually become less useful because a very large number of the I functions must be calculated. In the following we provide an assortment of useful formulas for this important regime. The expansions (A23) and (A24) provide accurate numerical values for much smaller values of τ than can be obtained using the asymptotic expansion (A27), at least without invoking specialized acceleration methods.

a. w>0, real

Suppose first that w is positive and real. Our starting point is the integral representation (A9). Now, if $\tau>4$, for all values of the integration variable we may approximate each of I_0 and I_1 by $e^x/(2\pi x)^{1/2}$, which is the leading term of each function's asymptotic expansion, [Eq. (13.5.1) of Ref. [46]]. Substituting in (A9) and using Eq. (7.4.7) of Ref. [46], we obtain

$$F(w,\tau)\sim\frac{1}{2w^{1/2}}\,e^{2\tau w}\mathrm{erfc}(2\tau w)^{1/2}\quad(\tau>4), \tag{A23}$$

where erfc denotes the complementary error function. Higher-order terms of the asymptotic expansion of I_0 and I_1 give rise to two distinct sets of terms, the first of which are of order $1/(2\tau)$ times the result (A23), while the second are of order $\exp(-2\tau)$. Both sets of terms may be ignored if $\tau>4$. This expansion is used in Sec. IV B 1.

b. w<−1, real

Results similar to (A23) can be given for the regime w real and $w<-1$. In this regime we use the integral representation (A7) and note that the exponential factor $e^{2\tau w}$ decays, while there is a contribution to the integral which grows exponentially, arising from values of the integration variable in the immediate vicinity of the upper limit τ. For these values of x we may again approximate the functions I_0 and I_1 as in the preceding paragraph. The dominant contribution to the integral can be obtained by integrating by parts and retaining the leading term. One readily finds that the exact result (A7) can be approximated by

$$F(w,\tau)\sim\frac{-1}{2(2\pi\tau)^{1/2}|w|}\quad(2\tau\gg 1). \tag{A24}$$

This result is utilized in Sec. IV B 2.

c. Asymptotic expansion

The complete asymptotic expansion of (A1) in powers of $1/\tau$ can be obtained by invoking the standard method of Laplace [54]. One substitutes in (A1) the Taylor series expansion of $(1-x)^{1/2}/(x+w)$ in powers of x, extends the upper limit of integration to ∞, and integrates term by term. Now

$$(1-x)^{1/2}/(x+w)=\frac{1}{w}\sum_{n=0}^{\infty}(-x/w)^n\sum_{k=0}^{n}\left(-\frac{1}{2}\right)_k\frac{(-w)^k}{k!}. \tag{A25}$$

One readily obtains as the final result

$$F(w,\tau)\sim\frac{1}{2w(2\pi\tau)^{1/2}}\sum_{n=0}^{\infty}\left(\frac{-1}{2w\tau}\right)^n D_n(w), \tag{A26}$$

where the expansion coefficient $D_n(w)$ is a polynomial in w given by

$$D_n(w)=\frac{(2n)!}{2^{2n}n!}\sum_{k=0}^{n}(-1/2)_k w^k/k!. \tag{A27}$$

APPENDIX B: GLAUBER KINETIC ISING MODEL

In this appendix we derive selected formulas pertaining to $S(q,t)$ for the one-dimensional Glauber kinetic Ising model. We have already developed the necessary formalism in Secs. II and III, and it is relatively straightforward to provide a parallel derivation for the Glauber model. We restrict our attention here to providing basic formulas. Numerical results and major asymptotic properties will be presented elsewhere [50].

In the Glauber kinetic Ising model, the allowed transitions are single-spin flips, $\sigma_i\to\sigma_i'=-\sigma_i$, and the basic form of the spin-flip master equation operator is given by [see discussion around (2.15)],

$$D^{SF}[\sigma|\sigma']=\sum_i\delta^{[i]}_{\sigma,\sigma'}D^i_{SF}[\sigma|\sigma']. \tag{B1}$$

Corresponding to single-spin flips, the local operator has the form [compare with (2.17)],

$$D^i_{SF}[\sigma|\sigma']=W_i^G(\sigma')(\delta_{\sigma_i',-\sigma_i}-\delta\sigma_i',\sigma_i)=-W_i^G(\sigma')\sigma_i\sigma_i', \tag{B2}$$

where $W_i^G(\sigma)$, the Glauber transition probability function [12], is constructed to satisfy detailed balance and is given by [compare with (2.20)],

$$W_i^G(\sigma)=\frac{\alpha}{2}\left(1-\frac{\gamma_F}{2}\,\sigma_i(\sigma_{i+1}+\sigma_{i-1})\right). \tag{B3}$$

In this appendix the quantity α denotes the spin-*flip* rate for uncoupled spins (distinct from the Kawasaki spin-exchange rate), and is taken as a phenomenological parameter of the model; the overall factor of two in (B3) is introduced for convenience. Note from (B3) that for ferromagnetic couplings the spin-flip rate is maximized when neighboring spins have values opposite to that at site i, $\sigma_{i\pm1}=-\sigma_i$,

while $W_i^G(\sigma)$ is minimized when $\sigma_{i\pm1}=\sigma_i$. For this reason, the Glauber model provides a relatively simple framework to study the dynamics of cooperative phenomena.

Combining (B1), (B2), and (B3), the single-spin equation of motion can be derived [compare with (2.21)],

$$\widetilde{D}_\sigma^{SF}\sigma_i=-2W_i^G(\sigma)\sigma_i=-\alpha\left(\sigma_i-\frac{\gamma_F}{2}(\sigma_{i+1}+\sigma_{i-1})\right). \tag{B4}$$

In the Glauber model, the single-spin equation of motion includes *only* single spins and does not involve higher-order spin terms, as is the case with the Kawasaki model (2.23). Fourier transforming (B4) [see (2.4)] thus diagonalizes the dynamical problem,

$$\widetilde{D}_\sigma^{SF}\sigma(q)=-\alpha(1-\gamma_F\cos q)\sigma(q)\equiv-\lambda(q)\sigma(q). \tag{B5}$$

Note that, in contrast to the Kawasaki model, the spectrum of relaxation rates $\lambda(q)$ is bounded between $\alpha(1+\gamma_F)$ and $\alpha(1-\gamma_F)$. In particular, $\lambda(q)$ does not vanish for any nonzero temperature, i.e., there is no conserved mode for this dynamical model. We remark that whereas $\sigma(q)$ is an eigenvector of the spin-flip operator for all temperatures, $\sigma(q)$ is an eigenvector of the spin-exchange operator only for $T_F=\infty$; see (2.24). For this reason the exact form of $S(q,t)$ can be established for the Glauber model for any final temperature, T_F, but only for $T_F=\infty$ in the case of the Kawasaki model.

The corresponding two-spin equation of motion for the Glauber model can be derived using (B1–B3) and is given by

$$\widetilde{D}_\sigma^{SF}\sigma_i\sigma_j=\sigma_i\widetilde{D}_\sigma^{SF}\sigma_j+\sigma_j\widetilde{D}_\sigma^{SF}\sigma_i+4\delta_{i,j}W_i^G(\sigma), \tag{B6}$$

which should be contrasted with (2.22). Taking the Fourier transform and proceeding as in Sec. II, we arrive at the equation of motion,

$$\frac{\partial S(q,t)}{\partial t}=-2\{\lambda(q)[S(q,t)-\chi(q,T_F)]+\alpha\gamma_F G(t)\}, \tag{B7}$$

where $G(t)\equiv\Phi_1(t)-u_F$, with $\Phi_1(t)$ the nearest-neighbor nonequilibrium correlation function, now for spin-flip dynamics. As mentioned in Sec. II C, and as we see here explicitly, the equation of motion for $S(q,t)$, since it includes only two-spin nonequilibrium correlation functions, does not entail an infinite hierarchy of associated equations of motion for higher-order correlation functions.

Of course, (B7) must be solved subject to the constraint of the sum rule (2.9) which arises solely because of the Ising fixed-length spin condition and is independent of the dynamical model. We can impose (2.9) on (B7) by requiring that the function $G(t)$ obey the constraint analogous to (3.2),

$$G(t)=-\frac{1}{2\pi\alpha\gamma_F}\int_{-\pi}^{\pi}dq\ \lambda(q)[S(q,t)-\chi(q,T_F)]. \tag{B8}$$

Just as with the Kawasaki model, the role of the sum rule is to effectively cause the equation of motion for $S(q,t)$ to be nonlocal in the q space [see discussion around (3.2)]. The formal solution to (B7) is then similar to (3.5) with $\Omega(q)$ replaced with $\lambda(q)$ and with

$$J(q,t)=2\alpha\gamma_F\int_0^t dt'\ G(t')\exp[-2\lambda(q)(t-t')]. \tag{B9}$$

As in Sec. III, the effect of imposing the sum rule is to require that $G(t)$ obey the integral equation (3.7), where the analogous quantities for the Glauber model are given by

$$P(t)=2\alpha\gamma_F\int_{-\pi}^{\pi}dq\ \exp[-2\lambda(q)t], \tag{B10}$$

and

$$Q(t)=\int_{-\pi}^{\pi}dq[S(q,0)-\chi(q,T_F)]\exp[-2\lambda(q)t]. \tag{B11}$$

Note that (B11) is formally identical to (3.9), except that the relaxation spectrum $\lambda(q)$ differs from that for the Kawasaki model.

It is thus clear that one can follow the steps presented in Sec. III and derive the exact solution for $S(q,t)$. The equations presented here serve as the starting point for a detailed analysis of $S(q,t)$, an analysis that we present elsewhere [50]. A key result of that analysis is that the solution can be expressed in terms of the dispersion integral $F(w,\tau)$, and hence it is straightforward to derive the major asymptotic properties of $S(q,t)$ for the Glauber model using the results of Appendix A. The purpose of the development we have provided here is to show the close formal similarity between the Glauber and Kawasaki models, even though the dynamics differ qualitatively in the lack of a conserved mode in the case of the Glauber model.

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[30] The origin of (2.35) is as follows. For the one-dimensional nearest-neighbor Ising model in a zero magnetic field, the equilibrium average of a product of an even number of spins factors into a product of two-spin averages, according to $\langle \sigma_{i_1}\sigma_{i_2}\cdots\sigma_{i_{2n}}\rangle = \langle \sigma_{i_1}\sigma_{i_2}\rangle\cdots\langle \sigma_{i_{2n-1}}\sigma_{i_{2n}}\rangle$, $n\geq 1$, where the indices are ordered such that $i_1\leq i_2\leq\cdots\leq i_{2n}$. For the four-spin products in (2.33), therefore, we have, in equilibrium and for $n\geq 1$, $\langle \sigma_{-1}\sigma_0\sigma_1\sigma_n\rangle = \langle \sigma_{-1}\sigma_0\rangle\langle \sigma_1\sigma_n\rangle = u\times u^{n-1} = u^n = \langle \sigma_0\sigma_n\rangle$.

[31] K. E. Shuler, Phys. Fluids **2**, 442 (1959).

[32] This scenario was explicitly demonstrated for the two-dimensional Glauber kinetic Ising model in Ref. [7], where, using renormalization group methods, it was shown that the strongly nonequilibrium response to a large and sudden change in magnetic field strength goes over to the predictions of linear-response theory in the long-time limit.

[33] See, for example, G. F. Mazenko and O. T. Valls, Phys. Rev. B **24**, 1419 (1981).

[34] We note that for infinite temperature, it is not strictly necessary to find the memory function; one can solve the equation of motion (2.24) directly in this limit to give (2.43).

[35] E. W. Montroll, J. Math. and Phys. **25**, 37 (1946).

[36] The dynamic critical exponent is given by $z=5$ for a conserved variable in one dimension; see, for example, J. H. Luscombe, Phys. Rev. B **36**, 501 (1987).

[37] J. S. Langer, M. Bar-on, and H. D. Miller, Phys. Rev. A **11**, 1417 (1975).

[38] The notion that nonequilibrium probability distribution functions can be treated as if they were the equilibrium distribution function, except characterized by a time-dependent temperature, has been termed ''canonical invariance'' by H. C. Andersen, I. Oppenheim, K. E. Shuler, and G. H. Weiss, J. Math. Phys. **5**, 522 (1964).

[39] The quantity $W(q,t)$ may, however, contribute to higher-order sum rules.

[40] See, for example, F. G. Tricomi, *Integral Equations* (Interscience, New York, 1957).

[41] The procedure we describe actually leads to the integral equation $2\pi G(t) = Q'(t) - \int_0^t d\tau P'(t-\tau)G(\tau)$, which is fully equivalent to (3.7).

[42] See, for example, E. T. Whittaker and G. N. Watson, *A Course of Modern Analysis*, 4th ed. (Cambridge University Press, Cambridge, 1927), p. 65.

[43] K. Binder, Phys. Rev. B **15**, 4425 (1977).

[44] See, for example, G. Arfken, *Mathematical Methods for Physicists*, 3rd ed. (Academic, New York, 1985), Sec. 15.12.

[45] I. S. Gradshteyn and I. M. Ryzhik, *Tables of Integrals, Series, and Products*, 4th ed. (Academic, New York, 1965).

[46] *Handbook of Mathematical Functions*, edited by M. Abramowitz and I. A. Stegun (Dover, New York, 1965), Chap. 9.

[47] See, for example, M. Luban and J. H. Luscombe, Phys. Rev. B **35**, 9045 (1987).

[48] Such arguments relating the random walk of domain boundaries to one-dimensional dynamic critical exponents were first introduced by R. Cordery, S. Sarker, and J. Tobochnik, Phys. Rev. B **24**, 5402 (1981). These arguments have been extended to the critical dynamics of spins on fractal geometries by J. H. Luscombe and R. C. Desai, *ibid*. **32**, 488 (1985); and J. H. Luscombe, J. Phys. A **20**, 1299 (1987).

[49] J. H. Luscombe and M. Luban, Surf. Sci. (to be published).

[50] J. H. Luscombe and M. Luban (unpublished).

[51] N. B. Ninham, R. Nossal, and R. Zwanzig, J. Chem. Phys. **51**, 5028 (1969).

[52] To arrive at (A6) we have used Eq. (13.2.1) of Ref. [46] as well as the identity ${}_1F_1(1/2;2;-z)=e^{-z/2}[I_0(z/2)+I_1(z/2)]$ which follows upon using formulas in Sec. 13.4 of that reference.

[53] G. N. Watson, *A Treatise on the Theory of Bessel Functions*, 2nd ed. (Cambridge University Press, Cambridge, 1958).

[54] C. M. Bender and S. A. Orszag, *Advanced Mathematical Methods for Scientists and Engineers* (McGraw-Hill, New York, 1978), Sec. 6.4.