Hamiltonian approach to the kinetic Ising models

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In the framework of the quantum approach to stochastic dynamics, the master equations governing the temporal evolution of the kinetic Ising models (KIM) are cast in the form of imaginary-time Schrödinger equations with second-quantized Hermitian Hamiltonians. On the basis of the quantum formalism a classical evolution equation is derived for an effective time-dependent Ising-type Hamiltonian. The grand ensemble corresponding to the latter describes the statistics of the spin configurations evolving under the dynamics of the KIM. The latter approach was used to qualitatively explain the results of recent Monte Carlo simulations of the Kawasaki KIM.

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The kinetic Ising models (KIM) with spin-flip (Glauber) [1] and spin-exchange (Kawasaki) [2] dynamics were originally designed [3] to introduce in a mathematically simple way a nonequilibrium kinetics into the Ising model (IM) which, as is known, does not possess any inherent dynamics. The Markovian character of the stochastic master equation (ME) governing the dynamics of KIM makes them very convenient objects for the application of the Monte Carlo method [3]. Numerous studies performed with the use of this technique revealed that KIM, in spite of their simplicity, describe in a qualitatively correct manner a broad variety of phenomena found in real systems [4,5]. Among these, major attention attracted essentially nonequilibrium phenomena qualitatively different from the familiar quasiequilibrium relaxation processes. Because of their novelty and in the absence of an exhaustive theory, these phenomena are necessarily being discussed in terms of such rather intuitive concepts as nonequilibrium phase diagrams, nucleation, spinodal decomposition, the late-stage scaling, etc. [4,6]. This lack of exactness makes it rather difficult to introduce these notions into the rigorous framework of the Markov chains theory [7] which, besides, is not well adopted to specifically physical applications.

Therefore, there have been continuing efforts [3,8–16] to cast KIM in more appropriate terms of physical theories, namely, of quantum and statistical mechanics, in order to use their concepts and techniques to study the physics underlying KIM. In the quantum approach to stochastic dynamics [2,3,8–13,17–21] one exploits the formal analogy between the ME and the (imaginary-time) Schrödinger equation. A general conclusion that may be drawn from these studies is that whenever a ME can be reduced to a Schrödinger equation with a Hermitian Hamiltonian, sophisticated techniques of quantum theory successfully apply [3,8,11,13,19,20]. Another conclusion that has not yet been fully appreciated in the KIM context is that the use of the second-quantization representation of Refs. [9,10] greatly clarifies both physical and formal aspects of the theory [12,13,18,21].

The statistical approach [14–16] aims at reducing the statistics of the ensembles of the IM configurations evolving under the stochastic dynamics of KIM to the form of canonical ensembles with some effective Hamiltonians. At present this approach has been successful only in accounting either for the most probable configurations [14,15] or in the case of steady states [16].

The aim of the present paper is to present a unified quantum-mechanical approach based on the techniques developed mainly in Refs. [3,8,9–11] by giving a second-quantized Hermitian Hamiltonian representation of the KIM and to develop on this basis a purely classical formalism in which the statistical ensembles evolving according to the KIM dynamics have the form of grand ensembles with effective time-dependent Ising Hamiltonians.

To comprise a maximally broad class of the Ising-type models, we will consider the Hamiltonians having the form of the most general functions of the Ising variables (IV) $\sigma_i = \pm 1$ defined at some set $\{i\}$ of N sites that do not necessarily form a regular lattice:

$$\mathcal{H} = \sum_{\alpha} K_\alpha \sigma_\alpha. \tag{1}$$

Here the notation of the cluster algebra [15] is used with the variable σ_α corresponding to a cluster $\alpha \subseteq \{i\}$ being expressed through the IV as

$$\sigma_\alpha = \prod_{j \in \alpha} \sigma_j. \tag{2}$$

The summation in Eq. (1) is over all possible subsets of $\{i\}$, including the empty one $\alpha = \emptyset$ with $\sigma_\emptyset \equiv 1$. The coefficient $K_\emptyset$ may be used to normalize the canonical probability distribution $\rho(\mathcal{H}) = \exp(-\mathcal{H})$. (The factor $1/k_B T$ is assumed to be included into the parameters of $\mathcal{H}$.) It is to be noted that because $\mathcal{H}$ is the most general function of $\{\sigma_i\}$, any identity valid for $\mathcal{H}$ will be valid for any particular function of the IV. So we will repeatedly use $\mathcal{H}$ as a trial function to derive general formulas, while the Ising Hamiltonians governing the dynamics of the KIM will be marked by the superscript I.

In the quantum notation of Ref. [8] the ME governing temporal evolution of the ensemble $|t\rangle$ of the microscopic states of an IM has the form

$$\frac{d}{dt}|t\rangle = \mathbf{T}|t\rangle, \tag{3}$$

where the transition matrix $\mathbf{T}$ must satisfy the condition of the probability conservation and the detailed balance condition

$$\langle|\mathbf{T} = 0 \quad \text{and} \quad \mathbf{T}|\mathcal{H}^I\rangle = 0, \tag{4}$$

respectively. Here $|\rangle$ corresponds to the sum of all possible microscopic states of the IM taken with equal weights and, by definition [8], $|\mathcal{H}\rangle = \exp(-\mathcal{H})|\rangle$.

A second-quantization representation of the ME (3) [9,10] is most easily introduced through the lattice gas model to which the IM is equivalent. Introducing the operators of creation (b_i^+) and annihilation (b_i^-) of the lattice gas atoms we can map the IM configurations onto the Fock space vectors by identifying the spins up (down) with the occupied (empty) states of the state vector

$$|\alpha\rangle = \prod_{i\in\alpha} b_i^+|\text{vac}\rangle, \tag{5}$$

where the vacuum state $|\text{vac}\rangle$ corresponds to the empty lattice (all spins down [15]) and vector $|\alpha\rangle$ corresponds to the configuration with spins in the cluster α pointing up. It is easy to see that with the help of $b_i^\pm$ the elementary processes in the KIM can be expressed through the Hermitian operators

$$H_i^{\text{in}} = -(b_i^+ + b_i^-) \tag{6a}$$

for spin flips and

$$H_{ij}^{\text{in}} = -(b_i^+ b_j^- + b_j^+ b_i^-) \tag{6b}$$

for spin exchanges [10] (other notation will be explained shortly). To make this picture fully consistent one has to ensure that each site i be occupied by not more than one atom, i.e., to satisfy the exclusion principle. This can be achieved by imposing upon $b_l^\pm$ the so-called hard-core boson or Pauli-type (anti)commutation relations [10] that can be realized through the algebra of spin-1/2 operators $\mathbf{s}_l$ [11–13,17,20–22]

$$b_l^\pm = s_l^\pm = s_l^x \pm i s_l^y, \tag{7a}$$

$$n_l = b_l^+ b_l^- = s_l^z + \tfrac{1}{2} = \tfrac{1}{2}(\sigma_l + 1). \tag{7b}$$

Below we will use both the boson and the spin operators, whichever is more convenient. To avoid repeated change of notation we will usually retain the original IV in all formulas by assuming that in a particular representation they have to be expressed through the appropriate operators according to Eq. (7b). In this formalism the state $|\rangle$ corresponds to the coherent state of the hard-core bosons [9,10]

$$|\rangle = \exp\left(\sum_i b_i^+\right)|\text{vac}\rangle = \prod_i (1 + b_i^+)|\text{vac}\rangle. \tag{8}$$

It was shown [8,7] that the Markovian generators $\mathbf{T}$ corresponding to the KIM are negative semidefinite operators. The results of Refs. [3,11,13] further suggest that $\mathbf{T}$ may be expressed through a Hermitian positive semidefinite Hamiltonian H as

$$\mathbf{T} = -\exp(-\tfrac{1}{2}\mathcal{H}^I) H \exp(\tfrac{1}{2}\mathcal{H}^I). \tag{9}$$

Indeed, let us define auxiliary Hamiltonians through the operators (6) as

$$H^{\text{in}} = \sum_\omega H_\omega^{\text{in}}.$$

Here and below the index ω and the term "ω process" correspond to the index i and the spin-flip process $\sigma_i \to -\sigma_i$ in the case of the Glauber dynamics, and to the index ij and the spin-exchange process $\sigma_i \rightleftharpoons \sigma_j$ in the case of the Kawasaki dynamics. Now it is the matter of a straightforward calculation with the use of the state vectors (5) to see that the matrix elements of the operator $\mathbf{T}^{\text{in}}$ defined according to Eq. (9) with $H = H^{\text{in}}$ have all the properties of the purely off-diagonal "scattering in" term defined in Ref. [8]. In particular, nonzero matrix elements of $\mathbf{T}^{\text{in}}$ correspond to the transition probabilities of the form

$$W(\alpha|\alpha_\omega) = \langle\alpha|\mathbf{T}^{\text{in}}|\alpha_\omega\rangle = \exp(-\delta_\omega \mathcal{E}^I/2), \tag{10}$$

where $\delta_\omega \mathcal{E}^I$ is the change of the energy (in units of $k_B T$) of a configuration α of the Ising spins under the ω process. This choice of W is used relatively rarely [3,11,15]. More usual ones can be obtained from Eqs. (6) and (9) by multiplying $\mathbf{T}_\omega^{\text{in}}$ with a function $\hat{W}_\omega$ of the IV, which is invariant with respect to the ω process: $\hat{W}_\omega(\{\sigma_i\}_\omega) = \hat{W}_\omega(\{\sigma_i\})$ [3] as, e.g., the function corresponding to the original choice of Glauber [1,3]

$$\hat{W}_j(\{\sigma_i\}) = \tfrac{1}{2}\text{sech}(\tfrac{1}{2}\delta_j \mathcal{H}^I) = \tfrac{1}{2}\text{sech}(E_j), \tag{11}$$

where $E_i = \text{tr}_i(\sigma_i \mathcal{H})/2$. For simplicity, in our general derivation we will continue to use the choice (10) because the generalization is straightforward. To complete the construction of the second-quantization representation of the ME we need to supplement the $\mathbf{T}^{\text{in}}$ term by the "scattering out" diagonal term $\mathbf{T}^{\text{out}}$: $\mathbf{T} = \mathbf{T}^{\text{in}} - \mathbf{T}^{\text{out}}$ [8] in order to satisfy the requirements (4). From Eq. (9) it follows that for H the two conditions unify to one [3]: $H|\frac{1}{2}\mathcal{H}^I\rangle = 0$, which gives us the equation for recovering H^{out},

$$H^{\text{in}}|\tfrac{1}{2}\mathcal{H}^I\rangle = H^{\text{out}}|\tfrac{1}{2}\mathcal{H}^I\rangle. \tag{12}$$

Below we sketch the derivation in more complicated spin-exchange case, the spin-flip case being treated similarly.

We first note that the diagonality of H^{out} can be ensured by the property of the operators $b_i^\pm$ to be equivalent to diagonal operators when acting at the coherent state (8)

$$b_i^\pm|\rangle = \tfrac{1}{2}(1 \pm \sigma_i)|\rangle. \tag{13}$$

So to find H^{out} we only need to commute H^{in} with $\exp(-\mathcal{H}^I/2)$ on the left-hand side (lhs) of Eq. (12). With

the use of the identity

$$\exp(\mathcal{H})b_i^{\pm}\exp(-\mathcal{H}) = \exp(\pm 2E_i)b_i^{\pm},$$

we find

$$b_i^{\pm}|\mathcal{H}\rangle = \tfrac{1}{2}(1 \pm \sigma_i)|\mathcal{H} \mp 2E_i\rangle. \tag{14}$$

Now using this relation for all four operators entering H_{ij}^{in} and the definition of $|\mathcal{H}\rangle$ we can express the lhs of Eq. (12) through the action of diagonal operators at $|\mathcal{H}\rangle$. To simplify the resulting expression it is helpful to use the algebra of the projection operators P_{ij}^{+} and $P_{ij}^{-} = 1 - P_{ij}^{+} = Q_{ij}^2$, where $Q_{ij} = (\sigma_i - \sigma_j)/2$. The final expression reads

$$H_{ij}^{\text{in}}|\mathcal{H}\rangle = [P_{ij}^{+} - \exp(-\delta_{ij}\mathcal{H})]|\mathcal{H}\rangle, \tag{15}$$

where use has been made of the general definition of the operator

$$\delta_\omega \mathcal{H} = \mathcal{H}_\omega - \mathcal{H},$$

which is antisymmetric under the ω process

$$(\delta_\omega)_\omega = -\delta_\omega. \tag{16}$$

Now unifying Eqs. (15), (12), (6b), and (7) we finally obtain for the case of the Kawasaki dynamics

$$H = \sum_{\langle ij\rangle}[-2\mathbf{s}_i \cdot \mathbf{s}_j - \tfrac{1}{2} + \exp(-\tfrac{1}{2}\delta_{ij}\mathcal{H}^I)], \tag{17a}$$

where the angular brackets denote the summation over some set of pairs of sites. Similarly, in the case of the Glauber dynamics the Hamiltonian has the form [11]

$$H = \sum_i [-2s_i^x + \exp(-\tfrac{1}{2}\delta_i \mathcal{H}^I)]. \tag{17b}$$

Thus, we have reduced the KIM to the Hamiltonian dynamics of quantum spin models (cf. Refs. [11–13,17,18,20]). This allows one to use the wealth of techniques developed in connection with these models to study the dynamics of KIM. For example, in the limit $T \to \infty$ $\delta_\omega \mathcal{H}^I = O(1/T) \to 0$ the Hamiltonians (17) reduce to the deposition-evaporation models considered in Ref. [20]. The exact results obtained in these papers for $T = \infty$ might be helpful in attacking the difficult problem [3] of developing a high-temperature expansion for the Kawasaki dynamics.

Despite the remarkable results of Ref. [20], the quantum spin models are, in general, very complicated ones which are difficult to solve even numerically [23]. Therefore, from the point of view of actual calculations the above formalism is not too advantageous. It is not even excluded that it could be more helpful the other way, i.e., in studying the spin models in the stochastic representation. For example, from Eq. (17a) it is easy to see that using the above $T = \infty$ limit one can express the partition function of the isotropic Heisenberg spin-1/2 model through the sum of diagonal matrix elements of the evolution operator $\exp(-tH)$. In other words, to reduce the statistics of quantum spins to the statistics of closed self-avoiding random walks on a lattice executed by an ensemble of identical particles.

In order to alleviate the difficulties involved in the quantum formalism it is natural to use, whenever possible, Eqs. (13) and (14) to replace nondiagonal operators $b_i^{\pm}$ by much simpler diagonal ones σ_i. The remarkable fact is that in some important cases the quantum operators can be discarded altogether. For example, using Eq. (14) it is easy to see that the state vector $|t\rangle$ may be represented in the form $|\mathcal{H}_t\rangle$ with some effective time-dependent Ising Hamiltonian $\mathcal{H}_t$. Substituting this into the ME (3) with $\mathbf{T}$ given by Eq. (9) and using Eq. (15) (and its analog for H_i^{in}) we arrive at the following equation for $\mathcal{H}_t$:

$$\dot{\mathcal{H}}_t = \sum_\omega [\exp(-\tfrac{1}{2}\delta_\omega \mathcal{H}^I) - \exp(\tfrac{1}{2}\delta_\omega \mathcal{H}^I - \delta_\omega \mathcal{H}_t)]. \tag{18}$$

This equation is the main result of the present paper. With the use of the effective Ising-type Hamiltonian $\mathcal{H}_t$ the problem of temporal evolution of multispin correlation functions is reduced to the conventional problem of finding the equilibrium average

$$\mu_\alpha(t) = \operatorname{tr}(\sigma_\alpha e^{-\mathcal{H}_t}) \equiv \langle\langle \sigma_\alpha \rangle\rangle_t. \tag{19}$$

To establish contact with more conventional approaches to the KIM, let us derive the evolution equation for $\mu_\alpha(t)$ in the case of the Glauber model. To this end we first introduce the factor (11) under the summation sign on the right-hand side (rhs) of Eq. (18). Then we multiply both sides of the equation by $\sigma_\alpha \exp(-\mathcal{H}_t)$ and take trace over $\{\sigma_i\}$. Next using the invariance of the trace under the reversal of spins, we make the change $\sigma_j \to -\sigma_j$ in the second term on the rhs with $\omega = j$. This leads to the cancellation between the two terms unless $j \in \alpha$. The remaining terms can be rearranged to give

$$\dot{\mu}_\alpha(t) = -\mu_\alpha(t) - \sum_{j\in\alpha}\langle\langle \sigma_j \sigma_\alpha \tanh(E_j)\rangle\rangle_t \ ,$$

which coincides with the equation obtainable directly from the ME [see, e.g., Eq. (3.42) in Ref. [3]]. Because the cluster variables (2) constitute a complete set in the phase space of the IM, this proves the equivalence of the theory given by Eq. (18) to the conventional ME approach to the KIM in the case of the one-time averages.

To make the above formalism efficient it is necessary to develop the methods of solving Eq. (18). As a first step in this direction let us consider the practically important case of initial ensembles corresponding to the quench from $T = \infty$. In this case a natural choice is the high-temperature expansion because, as is easily verified, it linearizes Eq. (18) to each order in $1/T$. To the order $1/T$

$$\dot{\mathcal{H}}_t \approx \sum_\omega (\delta_\omega \mathcal{H}_t - \delta_\omega \mathcal{H}^I). \tag{20}$$

To assess qualitative features of this approximation, let us apply it to the problem of the nearest neighbor (NN)

spin-exchange kinetics of a model equiatomic binary alloy on a d-dimensional hypercubic lattice with effective pair interaction $V^I_{mn} = 4k_BTK^I_{mn}$:

$$\mathcal{H}^I = \sum_{\langle mn \rangle} K^I_{mn}\sigma_m\sigma_n.$$

This form of the interaction is preserved by the linear approximation (LA). The evolution equation (20) for the effective pair interaction K_{mn} with the use of Fourier transform can be reduced to

$$\dot{K}_t(k) = -\epsilon_k[K_t(k) + K_t - K^I(k) - K^I]. \tag{21}$$

Here and everywhere below k is the wave vector,

$$\epsilon_k = 8\sum_{l=1}^{d} \sin^2(k_l/2), \tag{22}$$

$K_t(K^I)$ is the NN component of the matrix $K_{mn}(K^I_{mn})$, and the initial condition is $K_0(k) = 0$. Equation (21) is easily solved by the use of Laplace transform. A leading term of the solution that correctly reproduces its salient features has the form

$$K_t(k) \approx K^I(k)[1 - \exp(-\epsilon_k t)]. \tag{23}$$

Let us use this expression to discuss the applicability of the LA to the extensively studied problems (see Refs. [4,6,24], and references therein) of the kinetics of the IM quenched below its phase transition point. From Eq. (23) it follows that starting from $K_0(k) = 0$ at $t = 0$, $K_t(k)$ regains its asymptotic value $K^I(k)$ as $t \to \infty$. Therefore, at some finite time t_c the system must attain the point of phase instability. Thus, the LA can be valid only for $t < t_c$ because it is based on the high-temperature expansion which, as is known, fails at phase transition points. From mean-field-type arguments it follows that the instability will take place as the minimum of $K_t(k_m)$ at some k_m will reach a critical value K_c. The nature of the instability will depend on a particular form of $K^I(k)$. For example, if the transition is of the ordering type, the instability points are at some high-k values $\{k_m\}$. From Eqs. (23) and (22) it follows that at large k $K_t(k)$ recovers much more quickly than at low-k ones. So in the case of ordering the linear regime is restricted to very short time t_c, which is shortening quickly as the quenching temperature lowers. Besides, the breakdown of the LA begins at large $|k|$. These conclusions are in complete qualitative agreement with recent Monte Carlo simulations [25]. If the minima of $K^I(k)$ at high-k points are only local ones with the global minimum being at the origin, these local minima during a certain period of the early stage of evolution became the global minima of $K_t(k)$. This conclusion is qualitatively confirmed by the transient ordering observed in numerical simulations [24].

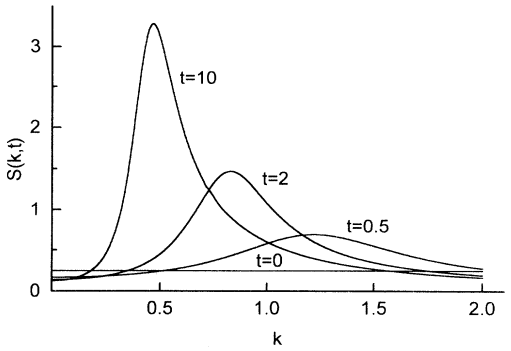

FIG. 1. Angle averaged structure factor $S(k,t)$ vs k (in units of inverse lattice spacing) at early stages of decomposition of a model binary equiatomic square alloy, with NN effective pair interaction $K^I = -0.5$.

To assess numerical values of the quantities under discussion, let us consider a two-dimensional alloy with NN interaction $K^I = -0.5$, which corresponds to quenching into the two-phase region slightly below the critical point $K^I_c \approx -0.44$:

$$K^I(k) = -(\cos k_x + \cos k_y). \tag{24}$$

From Eq. (23) it follows that $K_t(0) = 0$ for all t. So the minimum of $K^I(k)$ (24) at $k = 0$ splits into several minima of $K_t(k)$ at $\{k_m(t)\}$, which tend to zero as $t \to \infty$. These minima define the maximums of the experimentally observable structure factor $S(k,t) = \frac{1}{4}\langle\langle\sigma_{-k}\sigma_k\rangle\rangle_t$. The results of the calculation of this quantity exhibiting characteristic coarse-graining behavior [4] are shown in Fig. 1. $K_t(k)$ was calculated by the numerical integration of its Laplace transform; the statistical average has been performed with the use of the properly normalized [26] Krivoglaz-Clapp-Moss formula, which is very accurate at the equiatomic composition in the disordered phase [27]. The largest $t = 10$ in Fig. 1 approximately corresponds to t_c estimated from the condition $K_t(k_m) = K^I_c$. In the ordering case t_c would be almost two orders of magnitude smaller, which makes the linear regime practically unobservable [25].

In conclusion a word of caution is to be said about the above calculations in the two-phase region. As is known [3,7], in this case the dynamics of the KIM becomes nonergodic. Therefore, the conclusions drawn on the basis of the grand ensemble formalism used throughout the present paper may turn out not to be directly applicable to the interpretation of the data obtained, as is frequently the case in experiments (actual or numerical), for a particular specimen of the system under consideration. This, as well as other problems connected with nonergodic behavior of KIM require further investigation.

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