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# Real-Space Renormalization Group for Critical Dynamics

Ya'akov Achiam
*Physics Department, Queens University, Kingston, Ontario K7L 3N6, Canada*

and

J. M. Kosterlitz[a]
*Physics Department, Princeton University, Princeton, New Jersey 08540*

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We propose a real-space renormalization-group technique to treat dynamic critical phenomena in two-dimensional Glauber-Ising models. The invariant form of the time-dependent probability distribution is found within linear response theory and an estimate of the dynamic exponent $z$ is given.

In recent years much effort has been expended in extending the renormalization-group (RG) approach to critical dynamics.[1-5] This has been done by several methods based principally on expansions about two[2] and four dimensions[1,3,4] using a generalized Langevin equation of motion,[1,4] equivalent Lagrangian methods,[3] or the Fokker-Planck equation of motion.[5] All the above methods rely in an essential way on the separation into slow and fast modes which can be performed in continuous-spin models in momentum space. Up to now very little work has been done with discrete-spin models in real space and there is no systematic approach which gives reasonable agreement with known results in two dimensions.[6]

In this Letter, we present a general formulation of a real-space RG[7,8] to treat the critical dynamics of such discrete-spin models. Our method is an analytic one capable of dealing in a systematic manner with a multiplicity of time scales which appear in transient and memory effects in contrast to previous attempts to formulate a real-space RG. There is uncertainty[6,9,10] in the value of the dynamic exponent[11] $z$ even in the simplest model of purely dissipative dynamics of the two-dimensional Ising model. Since the real-space RG can give rather accurate results for the static critical behavior,[7] we believe that our method is potentially capable of comparable accuracy for the dynamics. The best result we have obtained to date is for the Glauber-Ising model[12] in two dimensions within the second-order cumulant expansion[7] on a triangular lattice: $z = 2.19$. This lies within the range of previous estimates by high-temperature expansions[9] and Monte Carlo methods[10] and is a considerable improvement over the value obtained in Ref. 6, which does not obey the inequality $z \geq 2 - \eta$.[13] Further results obtained using the method described below are (i) the exponent for the decay of energylike perturbations, $z_E \simeq 2.5$ (first-order cumulant approximation), and (ii) a rederivation of the exact results in one dimension, $z = 2$.[12,14] A fuller account will be presented in future publications and we can see no insuperable difficulty in using the method to extend other formulations of the static RG to critical dynamics and to study more complicated dynamics with conservation laws[13]

The equilibrium properties of the two-dimensional Ising model are determined by a reduced Hamiltonian[7] $\bar{H} = -H/kT = \sum K_a S_a(\sigma)$, where the $S_a(\sigma)$ are extensive functions of subsets of the spins $\sigma_i = \pm 1$. The dynamical model we study is that proposed by Glauber[12] in which the spins are assumed to flip independently in time $\tau$. The kinetic equation obeyed by the probability distribution $P(\sigma, t)$ is

$$\tau dP(\sigma, t)/dt = -L(\sigma)P(\sigma, t) = -\sum_j (1-p)W_j(\sigma_j)P(\sigma, t), \quad (1)$$

where the operator $P_j$ flips the spin $\sigma_j$, $p_j f(\sigma_j) = f(-\sigma_j)$. The transition probabilities $W_j(\sigma_j)$ are subject to the condition of detailed balance which ensures that, as $t \to \infty$, the probability distribution reaches its equilibrium form $P_e(\sigma) = \exp\{\bar{H}\}/Z$, where $Z$ is the partition function. We choose the form

$$W_j(\sigma_j) = [P_e(-\sigma_j)/P_e(\sigma_j)]^{1/2}. \quad (2)$$

If the Hamiltonian contains only nearest-neighbor interactions,

$$\bar{H} = K_2 \sum_{<ij>} \sigma_i \sigma_j,$$

then $W_j(\sigma_j) = \exp\{-K_2 \sigma_j \sum_i \sigma_i\}$, where $i$ and $j$ are nearest neighbors.[13] One could consider other forms of $W$, but it can be shown that the only difference is in transient and memory effects and will not affect the dynamic exponent $z$. This particular choice of $W$ eliminates all such problems up to first order in the cumulant approximation. They first appear at second order.

We consider small deviations from equilibrium within linear response theory, described by a function $\varphi(\sigma, t) = P(\sigma, t)/P_e(\sigma)$, where at $t = 0$, $\varphi(\sigma, 0) = 1 + h \sum \sigma_i$. The probability distribution can be parametrized by a time-dependent Hamiltonian $P(\sigma, t) \equiv \exp\{\bar{H}(\sigma, t)\}/Z(t)$ where, within linear response, $Z(t) = Z + O(h^2)$. Thus, the normalization of $P(\sigma, t)$ is independent of time and can be ignored in the rest of the discussion. The time-dependent Hamiltonian can be parametrized in exactly the same way as the equilibrium Hamiltonian except that the coefficients $K_a$ will be functions of time. They can, in principle, be determined from the master equation (1) with a given initial condition. Within linear response, the function $\varphi(\sigma, t)$ may also be parametrized in the form $1 + h_1(t) \sum \sigma_i + h_3(t) \sum \sigma_i \sigma_j \sigma_k + \ldots \equiv 1 + \vec{O}(\sigma) \circ \vec{h}(t)$.

The renormalization-group transformation is a standard static one defined by

$$\exp\{\bar{H}'(\mu)\} = \sum_\sigma T(\mu, \sigma) \exp\{\bar{H}(\sigma)\} \quad (3)$$

with a suitable choice of $T(\mu, \sigma)$.[7] Since the time-dependent Hamiltonian $\bar{H}(\sigma, t)$ contains the time $t$ as the corresponding equilibrium ones. However, the properties of the master equation (1) under the RG will determine how the characteristic time $\tau$ scales. Equation (1), which can be written as $\tau(d/dt)\vec{O}(\sigma) \cdot \vec{h}(t) = -\mathfrak{L}(\sigma)\vec{O}(\sigma) \cdot \vec{h}(t)$ with $\mathfrak{L}(\sigma) = \sum_i W_i(\sigma_L)(1 - p_i)$, is transformed under the RG to

$$\tau(d/dt)\vec{O}(\mu) \cdot \underline{\Lambda}\vec{h}(t) = -\mathfrak{L}'(\mu)\vec{O}(\mu) \circ \underline{\Omega}\vec{h}(t), \quad (4)$$

where $\underline{\Lambda}$ and $\underline{\Omega}$ are matrices which are defined by the RG transformation. If $\underline{\Lambda}$ and $\underline{\Omega}$ commute, the eigenvector of $\underline{\Lambda}\underline{\Omega}^{-1}$ with the largest eigenvalue will describe the slowest mode. However, in general they will not commute and then one must apply the RG transformation a large number of times. We then see that the slowest mode will scale with an exponent $z$ given by $b^z = \lambda/\omega$, where $\lambda$ ($\omega$) are the largest eigenvalues of $\underline{\Lambda}$ ($\underline{\Omega}$) and $b$ is the space rescaling factor. Since it can be readily seen that higher derivatives in Eq. (1) decay rapidly under the RG, this noncommutivity of $\underline{\Lambda}$ and $\underline{\Omega}$, with $W$ given by Eq. (2), is the only remaining memory effect and first appears at second order.

We will now put these ideas into practice using as an illustration the RG on a square lattice in the first cumulant approximation. This example has the advantage of demonstrating the essential ideas but avoids the technical complications met at higher orders and with different choices of $W(\sigma)$.

Since the $W_j(\sigma_j)$ are determined by $\bar{H}(\sigma)$ in Eq. (2), one can make a consistent approximation of both the Hamiltonian the time evolution operator $L(\sigma)$. Unfortunately, this approximation leads to rather poor results because $\varphi(\sigma, t)$ is an odd perturbation. It is well known that the first-order cumulant approximation gives negative values for the magnetic exponent $\beta/\nu$.[7,15] We find a dynamic exponent[1,4,11] $z \simeq 2.7$ which is rather higher than other estimates, but there is a great improvement at second order. However, the kinetic exponent[16] $\delta_k = z - 2 + \eta \simeq 0.4$, which is a measure of the deviation from the conventional theory,[17] is more in line with other estimates[6,9,10] which lie in the range $0.1 < \delta_k < 0.6$. At second order we obtain $\delta_k = 0.49$.

The renormalization-group transformation is defined by choosing $T(\mu, \sigma)$ in Eq. (3) by[8]

$$T(\mu, \sigma) = \prod_\alpha [1 + \mu_\alpha t_\alpha(\sigma)]/2 \equiv \prod_\alpha T_\alpha, \quad (5)$$

where the product is over the four-spin cells labeled $\alpha$ and $\mu_\alpha = \pm 1$ is the cell spin (see Fig. 1).

We take

$$t_\alpha(\sigma) = \sum_{i \in \alpha} \sigma_i (3 - \prod_{j \in \alpha} \sigma_i)/8 \quad (6)$$

following the suggestion of Barger[15] who showed that, within the first-order cumulant approximation, this form yields the lowest upper bound for the free energy. Let us now assume that the relevant part of the function $\varphi(\sigma, t)$ can be written as $1 + h(t) \sum \sigma_i$. This implies that, at time $t$, the system is in thermal equilibrium with an effec-

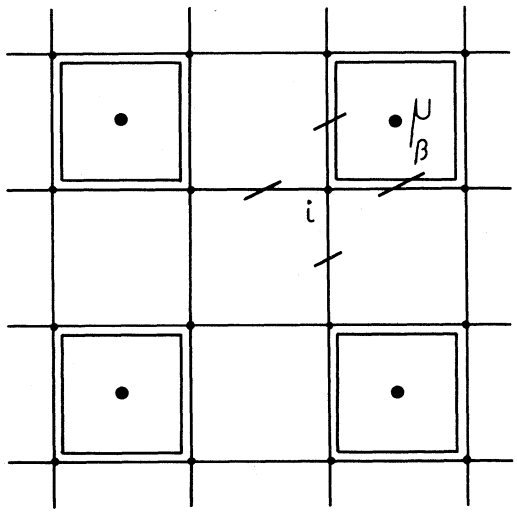


FIG. 1. Spin and cell lattice. Double (single) lines denote intracell (intercell) interactions in $P_\alpha{}^0(V)$. The slashes denote omitted interactions in $P_\beta{}^{(i)}$ and $V_\beta{}^{(i)}$.

tive field $h(t)$ whose time evolution is determined by Eq. (1). We will show later that $\sum\sigma_i$ is the only relevant operator in our approximation.

The RG transformation of Eq. (1) is now straightforward. Suppressing normalization factors, we write the equilibrium probability distribution $P_e(\sigma)=\prod_\alpha P_\alpha{}^0(\sigma)\exp\{V(\sigma)\}$, where $P_\alpha{}^0$ is the intracell part and $\exp\{V(\sigma)\}$ the intercell part.[7] Applying the RG transformation we have

$$\tau\partial P'(\mu,t)/\partial t = 2h(t)\textstyle\sum_\sigma\prod_\alpha P_\alpha{}^0 T_\alpha \exp\{V\}\sum_j W_j\sigma_j, \quad (7)$$

where $P'(\mu,t)=\sum_\sigma T(\mu,\sigma)P(\sigma,t)$. We concentrate on a single spin $\sigma_i$, i.e., on a single term in the sum over $j$ in Eq. (7). Divide the intercell interaction into two parts $V=\overline{V}_\beta+V_\beta$, where $V_\beta$ is the interaction between the cell containing $\sigma_i$, which is labeled $\beta$, and its neighbors, and $\overline{V}_\beta$ is the rest. Using the properties of $W_i(\sigma_i)$ we have the relation

$$P_\beta{}^0 W_i\exp\{V_\beta\}=P_\beta{}^{(i)}\exp\{V_\beta{}^{(i)}\}, \quad (8)$$

where the superscript $(i)$ means that all bonds connected to site $i$ are omitted as in the figure. Now the only remaining dependence on $\sigma_i$ in the term under consideration is of the form $T_\beta\sigma_i$ so that, when we sum over $\sigma_i=\pm1$, the only surviving contribution is $t_\beta\mu_\beta\sigma_i/2$ and we obtain

$$h(t)\mu_\beta\textstyle\sum_\sigma\prod_\alpha{}' T_\alpha P_\alpha{}^0 t_\beta P_\beta{}^{(i)}\sigma_i\exp\{\overline{V}_\beta+V_\beta{}^{(i)}\}, \quad (9)$$

So far this is exact, but to compute the trace in (9) we turn to the first-order cumulant approximation[7] and find

$$h(t)\mu_\beta Z_0{}^N(L_1/Z_0)\exp\{\langle\overline{V}_\beta\rangle\}, \quad (10)$$

where

$$Z_0=\sum_{\sigma\in\alpha}T_\alpha P_\alpha{}^0, \quad L_1=\sum_\sigma\sum_\beta t_\beta P_\beta{}^{(i)},$$

$N$ is the number of cells, and $\langle\ldots\rangle$ denotes an average over the $\sigma$ with weight function $\prod_\alpha{}' T_\alpha P_\alpha{}^0$. All of the quantities appearing in the expression (10) are easily calculated. Note that the contribution from $V_\beta{}^{(i)}$ in (9) vanishes in this approximation by symmetry considerations. The average $\langle\overline{V}_\beta\rangle$ is precisely that appearing in the corresponding *static* RG and is the new cell interaction,

$$\langle\overline{V}_\beta\rangle=K'\textstyle\sum'\mu_\alpha\mu_\gamma, \quad (11)$$

where the sum is over all nearest-neighbor cells excluding the cell $\beta$ containing $\sigma_i$. However, using the cell analog of Eq. (8), we find

$$Z_0{}^N\exp\{\langle\overline{V}_\beta\rangle\}=P_e'(\mu)W_\beta'(\mu), \quad (12)$$

where $P_e'(\mu)$ is the new equilibrium probability distribution of the variables and $W_\beta'(\mu)$ the new transition probability obeying Eq. (2). Summing over the four spins in each cell we obtain the renormalized master equation

$$\tau\partial P'(\mu,t)/\partial t=-CL'(\mu)P'(\mu,t) \quad (13)$$

where $L'(\mu)$ is of exactly the same form as $L(\sigma)$ but with rescaled interaction parameter $K_2\to K_2'$. The constant $C=2h(t)L_1/h'(t)Z_0$ is independent of time because $h'(t)$ is the new magnetic field at fixed $t$ and is given in terms of $h(t)$ via *static* RG as $h'(t)/h(t)=\Lambda_h$.[7,15] In our formulation, the length is rescaled by a factor $b=2$ at each stage and the constant $C$ can be absorbed into a redefinition of the time scale $\tau\to\tau'=\tau/C$. The dynamical exponent is determined by $b^{-z}=C$ yielding $z\simeq2.7$.

Since our renormalization of the master equation (1) with an assumed deviation from equilibrium of the form $\varphi(\sigma,t)=1+h(t)\sum\sigma_i$ reproduced a new $\varphi(\mu,t)$ of exactly the same functional form, this verifies that this is the appropriate fixed-point form and confirms dynamic scaling in this approximation. In this example the matrices $\underline{\Lambda}$ and $\underline{\Omega}$ in Eq. (4) are scalars and there are no memory effects which have been eliminated by our choice of $W(\sigma)$. We have checked explicitly that terms which are created by the time evolu-

tion[14] of $P(\sigma, t)$ reduce after a single RG step either to the form assumed in this work or to terms of second order in the intercell interaction. Of course, if the RG is performed to higher orders in the cumulant expansion, more terms must be used in $\varphi(\sigma, t)$ and the effective time-dependent Hamiltonian and transition probability become more complicated than the simple nearest-neighbor form.

We have used this method in the second-order cumulant approximation and confirmed dynamic scaling to this order. The calculations are lengthy and memory effects unavoidable. Details will be published elsewhere.

[a]On leave from University of Birmingham, Birmingham B15 2TT, England.

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# Dynamics of Energy Transport in Molecular Crystals: The Picosecond Transient-Grating Method

J. R. Salcedo and A. E. Siegman
*Department of Electrical Engineering, Stanford University, Stanford, California 94305*

and

D. D. Dlott and M. D. Fayer
*Department of Chemistry, Stanford University, Stanford, California 94305*

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Energy transport in molecular solids is investigated using a picosecond transient-grating technique. A model is presented which directly relates the rate of energy transport to the experimental observable. The diffusion coefficient for singlet electronic excitation transport is reported for pentacene in *p*-terphenyl. Acoustically induced optical absorption effects are also observed.

Intermolecular interactions in molecular crystals give rise to electronic excited-state energy migration. In pure crystals energy transport may be excitonic[1] in nature, and in disorded or impure crystals it may be due to long-range resonant transfer.[2] In this paper we present the first results from a new method which utilizes a picosecond transient diffraction grating to directly probe the time-dependent position of excited states in the bulk of a crystal. As such, this method allows determination of the rate of energy transfer along any crystallographic direction, and thus the influence of intermolecular interactions,[1] molecular orientation,[1] impurity effects,[3] and exciton-phonon coupling[4] on the transport process can be directly studied.

In this experiment, two time-coincident, Gaussian picosecond excitation pulses of wavelength

 