- (2) I. Lee and I. S. Koo, Tetrahedron, in press.
- (3) M. J. Kamlet, J. L. M. Abboud and R. W. Taft, "Progress in Physical Organic Chemistry," Ed. by R. W. Taft, Vol. 13, p.485–630, John Willy & Sons, 1981.
- (4) M. H. Abraham, R. W. Taft and M. J. Kamlet, J. Org. Chem., 46, 3053 (1981).

(5) (a) S. Winstein, E, Grunwald and J. W. Jones, J. Amer. Chem. Soc., 73, 2700 (1951); (b) A. H. Fainberg and S. Winstein, ibid., 78, 2770 (1956); (c) D. N. Kevill and M. S. Bahari, J. C. S. Chem. Commun., 572 (1982); (d) F. L. schadt, T. W. Bently and P. V. R. Schleyer, J. Amer. Chem. Soc., 98, 7667 (1976).

The Critical Dynamics for the Two-Dimensional Two-Spin Exchange Ising Model

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Kawasaki1 has studied the diffusion constant of spins in ferromagnets or molecules in binary mixtures near the critical point with the aid of the terminology of the spin system. His model is a time-dependent Ising model in which spin interactions are replaced by certain temperature-dependent transition probabilities of spin exchange. The transition probability of spin exchange is so chosen as to give the same equilibrium spin distribution function as conventional Ising system. In this respect, his model is quite similar to Glauber's, but is different from his in that the total spins are conserved at each transition. Achiam and Kosterlitz² have proposed a real-space renormalization group (RG) technique to treat dynamic critical phenomena in two-dimensional Ising model, which is just the extension of the real-space static RG method developed by Niemeijer and van Leeuwen³ and Kadanoff and Houghton⁴. Their model is that proposed by Glauber in which only one spin flips independently at a time from one state to another according to the prescribed transition probability. In this letter, we extend the RG method in realspace to Kawasaki's Ising model with two spin exchange. Then, we obtain a dynamic exponent for energy fluctuations.

The system we shall consider is a homogeneous two-dimensional Ising system. Let us apply a weak external force to the system at equilibrium. Then, the system will relax towards the equilibrium through an interaction with a heat reservoir. It is assumed that only two spins exchange at a time during the relaxation. The probability $P(\sigma_j, \sigma_k, \{\sigma\}_{jk}, t)$ for the spin-exchange on which the pair of spins σ_j and σ_k on the sites j and k exchanges each other may be described as

$$\tau \frac{\partial}{\partial t} P(\sigma_j, \sigma_k, \{\sigma\}_{jk}, t)
= -\sum_{\langle jk \rangle} (1 - p_{jk}) W_{jk}(\sigma_j, \sigma_k, \{\sigma\}_{jk}) P(\sigma_j, \sigma_k, \{\sigma\}_{jk}, t)
= -L(\sigma) P(\sigma_j, \sigma_k, \{\sigma\}_{jk}, t),$$
(1)

where τ is the bare relaxation time, p_{jk} the operator to exchange σ_j and σ_k , $W_{jk}(\sigma_j, \sigma_k, \{\sigma\}_{jk})$ the transition probability in which the pair of σ_j and σ_k exchanges each other and $\{\sigma\}_{jk}$ denotes the set of spins except σ_j and σ_k .

It should be noted that the time-displacement operator, L, is Hermitian. As $t\rightarrow\infty$, P_{eq} $(\sigma_j, \sigma_k, \{\sigma\}_{jk})$ satisfies the condition of detailed balance

$$W_{jk}(\sigma_j, \sigma_k, \{\sigma\}_{jk}) P_{eq}(\sigma_j, \sigma_k, \{\sigma\}_{jk})$$

$$= W_{jk}(\sigma_k, \sigma_j, \{\sigma\}_{jk}) P_{eq}(\sigma_k, \sigma_j, \{\sigma\}_{jk})$$
(2)

One may choose any specific form of the transition probability which obeys the above condition. We take the similar form of the transition probability used by Achiam and Kosterlitz instead of Kawasaki

$$W_{jk}(\sigma_{j}, \sigma_{k}, \{\sigma\}_{jk}) = \{P_{eq}(\sigma_{k}, \sigma_{j}, \{\sigma\}_{jk})/P_{eq}(\sigma_{j}, \sigma_{k}, \{\sigma\}_{jk})\}^{1/2} \cdot \exp(H_{jk})$$
(3)

where H_{jk} is the Hamiltonian due to the products of σ_j and σ_k .

It can be seen that $W_{jk}(\sigma_j, \sigma_k, \{\sigma\}_{jk}) P_{eq}(\sigma_j, \sigma_k, \{\sigma\}_{jk})$ is independent of the spins σ_j and σ_k . The equilibrium probability distribution can be described as

$$P_{eq}(\sigma_j, \ \sigma_k, \ \{\sigma\}_{jk}) = \exp\left\{-H(\sigma)\right\}/Z;$$

$$H(\sigma) = K_2 \sum_{(n,n,\cdot)} \sigma_m \sigma_n + K_3 \sum_{(n,n,n,\cdot)} \sigma_{m'} \sigma_{n'} + \cdots$$
(4)

where (n.n.) and (n.n.n.) mean the nearest neighborhood and the next nearest neighborhood, respectively, and the coupling constants K_2 and K_3 are related with the true coupling constants J_2 and J_3 by $K_2=J_2/k_BT$ and $K_3=J_3/k_BT$. The probability distribution at time t can be parameterized by a time-dependent Hamiltonian through the coupling constants and the partition function at t can be replaced by the equilibrium partition function within the linear response. Applying the RG transformation matrix, $T(\mu, \sigma)$, used in the static case by Niemeijer and van Leeuwen³ and Kadanoff and Houghton⁴ to the probability distribution at t, we obtain the new distribution for the new set of spin variable $\mu(=\pm 1)$ on a lattice with a lattice constant larger by a scaling factor of t

$$P'(\mu, t) = \exp\{-H'(\mu, t)\}/Z$$

$$= \sum_{|\sigma = \pm 1|} T(\mu, \sigma) P(\sigma, t);$$

$$H'(\mu, t) = K_2'(t) \sum_{(n.n.)} \mu_{\alpha} \mu_{\beta} + K_3'(t) \sum_{(n.n.n)} \mu_{\nu} \mu_{\sigma} + \cdots, \quad (5)$$

where $K_2'(t)$ and $K_3'(t)$ are the time-dependent coupling constants between the nearest neighboring and the next nearest neighboring lattice spins, respectively, and the form of the matrix is given as^{3,4}

$$T(\mu, \sigma) = \prod T_{\alpha}(\mu, \sigma) = \prod 1/2(1 + \mu_{\alpha}t_{\alpha}(\sigma)). \tag{6}$$

From eq. (5) the renormalized equation can be obtained

$$\overrightarrow{K}'(t) = R\{\overrightarrow{K}(t)\}. \tag{7}$$

Now, let us expand $P(\sigma_j, \sigma_k, \{\sigma\}_{jk}, t)$ near the equilibrium as follows

$$P(\sigma_{j}, \sigma_{k}, \{\sigma\}_{jk}, t) / P_{eq}(\sigma_{j}, \sigma_{k}, \{\sigma\}_{jk}) \simeq 1 + O(\sigma) \cdot \overrightarrow{h}(t)$$

$$= 1 + \frac{1}{2} \sum_{\langle m, n \rangle} \{ E_{m}(t, \sigma) \sigma_{m} + E_{n}(t, \sigma) \sigma_{n} \}, \qquad (8)$$

where

$$E_m(t,\sigma) = h_2(t) \sum_{(n,n,\cdot)} \sigma_{m'} + h_3(t) \sum_{(n,n,n,\cdot)} \sigma_{n'} + \cdots$$
 (9)

Substituting eq. (8) into eq. (1), using the condition of detailed balance and applying the RG transformation matrix to the result, we obtain

$$\tau \frac{\partial}{\partial t} \{ O'(\mu) \cdot \Lambda \vec{h}(t) \} = -L'(\mu) O'(\mu) \cdot \Omega \vec{h}(t), \tag{10}$$

The matrix Λ is just the same as that defined in the static case^{3,4} and Ω does not, in general, commute with Λ . Let λ_T and ω be the largest relevant eigenvalues of Λ and Ω , respectively. Then, we may rewrite eq. (10) as

$$\tau' \frac{\partial}{\partial t} \{O'(\mu) \cdot \vec{h}(t)\} = -L'(\mu)O'(\mu) \cdot \vec{h}(t), \tag{11}$$

where the renormalized relaxation time defined as

$$\tau' = (\lambda_T/\omega)\tau = l^z\tau. \tag{12}$$

Here, z is a dynamic exponent defined as $\tau' \propto \xi^z$, ξ being the correlation length.

In order to calculate the dynamic exponent z, let us substitute eq. (7) into eq. (1) and use the condition of detailed balance. Then, eq. (1) may be written as

$$\tau \frac{\partial}{\partial t} P(\sigma_j, \sigma_k, \{\sigma\}_{jk}, t) = -\sum_{(jk)} W_{jk}(\sigma_j, \sigma_k, \{\sigma\}_{jk})$$

$$P_{eq}(\sigma_j, \sigma_k, \{\sigma\}_{jk}) \sigma_j \exp\{2E_j(\sigma, t\}, (13)\}$$

where use has been made of the fact that $\sigma_k = -\sigma_j$ and $E_k(\sigma, t) = -E_j(\sigma, t)$ only contribute to the spin exchange and the unimportant static part has been neglected. Now, let us assume that the two spins in β lattice exchange each other. The total Hamiltonian can be separated into four parts, that is, the Hamiltonian in the β lattice, $H_{0\beta}$, the sum of the other lattices, $\sum_{\beta \neq \alpha} H_{0\alpha}$, the interaction between the site spins in the β lattice and another lattice, V_{β} , and the interaction between the site spins in the different lattices except the β lattice, V'. Then, we may write

$$W_{jk}(\sigma_{j}, \sigma_{k}, \{\sigma\}_{jk}) P_{eq}(\sigma_{j}, \sigma_{k}, \{\sigma\}_{jk})$$

$$= \prod_{\alpha \neq \beta} P_{eq}{}^{\alpha}(\sigma) P_{eq}{}^{\beta(jk)}(\sigma) \exp\{V_{\beta}{}^{(jk)} + V'\}, \qquad (14)$$

where the superscript in $P_{eq}^{\beta(jk)}$ and $V_{\beta}^{(jk)}$ means that

the probability distribution and the interaction are independent of σ_j and σ_k . Assuming that the interaction between the site spins in the different lattices is the perturbed term as in the static case, substituting eq. (14) into eq. (13) and then applying the RG transformation matrix to the result, we obtain

$$\tau \frac{\partial}{\partial t} P'(\mu, t) = -\sum_{j} \sum_{(\sigma = \pm 1)} \prod_{\alpha \neq \beta} T_{\alpha} \bar{t}_{\beta} P_{eq}{}^{\alpha}(\sigma) P_{eq}{}^{\beta(jk)}(\sigma)$$

$$\mu_{\beta} \exp\{V_{\beta}{}^{(jk)} + V' + 2E_{j}(\sigma, t)\}, \tag{15}$$

where the superscript prime in the summation denotes that σ_j and σ_k have been omitted and the new RG transformation matrix for the β lattice is defined as

$$\bar{t}_{\beta} = \sum_{|\sigma| = \pm 1|} 1/2\sigma_j t_{\beta}(\mu, \sigma). \tag{16}$$

Defining the partition functionals in α lattice and β lattice as

$$Z_{0\alpha} = \sum_{(\sigma = \pm 1)\alpha} T_{\alpha}(\mu, \sigma) P_{eq}{}^{\alpha}(\sigma),$$

$$Z_{0\beta}{}^{(jk)} = \sum_{(\sigma = \pm 1)} \tilde{t}_{\beta}(\mu, \sigma) p_{eq}{}^{\beta(jk)}(\sigma),$$
(17)

eq. (15) may be rewritten as

$$\tau \frac{\partial}{\partial t} P'(\mu, t) = -\sum_{j} \mu_{\beta} Z_{0\alpha}^{N-1} Z_{0\beta}^{(jk)}$$

$$< \exp\{V' + V_{\beta}^{(jk)} + 2E_{j}(\sigma, t)\} >_{0}, \tag{18}$$

where N is the number of lattices and the zeroth order average of a physical quantity, A, over site spins is defined as

$$\langle A \rangle_0 = \sum_{(\sigma = \pm 1)'} \prod_{\alpha \neq \beta} T_{\alpha} \hat{t}_{\beta} P_{eq}{}^{\alpha}(\sigma) P_{eq}{}^{\beta(jk)}(\sigma) A / Z_{0\alpha}{}^{N-1} Z_{0\beta}{}^{(jk)}$$
(19)

We may write the new probability distribution of the lattice spins as

$$\tau \frac{\partial}{\partial t} P'(\mu, t) = -\mu_{\beta} W'_{\beta}(\mu) P_{\epsilon q'}(\mu) \exp\{\vec{E}'(\mu, t)\};$$

$$E'(\mu, t) = h'_{2}(t) \sum_{(n,n,t)} \mu_{\beta} + h'_{3}(t) \sum_{(n,n,n,t)} \mu_{\nu} + \cdots$$
(20)

From the result in the static case $^{2\sim4}$, $W'_{\beta}(\mu)P_{eq}'(\mu)=Z_{0\alpha}^{N}<\exp(V')>_{0}$ and using the cumulant approximation, $\vec{E}'(\mu,t)$ has the following relation with $\vec{E}(\sigma,t)$ within the linear response

$$\vec{E}'(\mu, t) = \sum_{j} \{2Z_{0\beta}^{(jk)}/Z_{0\alpha}\} \{\langle \vec{E}(\sigma, t) \rangle_{0} + \langle (V_{\beta}^{(jk)} + V')\vec{E}(\sigma, t) \rangle_{0} - \langle (V_{\beta}^{(jk)} + V') \rangle_{0} \langle \vec{E}(\sigma, t) \rangle_{0} + 1/2 \{\langle (V_{\beta}^{(jk)} + V')^{2}\vec{E}(\sigma, t) \rangle_{0} - \langle (V_{\beta}^{(jk)} + V')^{2} \rangle_{0} \langle \vec{E}(\sigma, t) \rangle_{0} \} + \cdots \}.$$
(21)

From the above relation we may calculate the eigenvalue of Ω explicitly with the aid of a specific lattice. To obtain z we have taken the triangular lattice with seven spins and the matrix as $t_{\alpha}(\sigma) = \tanh (s \sum_{i \neq j} \sigma_i^{\alpha})$, s being a parameter. It is obvious that the critical exponents do not depend on the parameter. We shall take the limit $s \to \infty$. In actual calculation the nearest interaction between spins has been considered. The detailed calculation is so complicated that we write down the result in this letter. The characteristic time exponent for the energy fluctuations, z, is about 1.98, taking the terms up to the second order cumulant approximation. Kawasaki¹ has obtained the critical exponent for

the spin relaxation time, $\Delta \approx 2.00$ for square lattice. Schneider⁶ has shown that the critical exponent for the spin relaxation time is the same as that of the energy relaxation time, using the two-dimensional one-spin-flip Ising model. His theory holds for the two-spin exchange model, since the time-displacement operator, L, is also Hermitian. The critical exponent for the energy relaxation time, Δ , is related with z through $\Delta = \nu z$, ν being the critical exponent for the correlation length. Taking $\nu = 1$ for the two-dimensional Ising model, the present result is in an excellent agreement with Kawasaki's.

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References

- K. Kawasaki, *Phys. Rev.* **145**, 224 (1966); K. Kawasaki, in "Phase Trnasition and Critical Phenomena", edited by C. Domb and M. S. Green, Academic, New York, Vol. 2, 443, 1972.
- Y. Achiam and J. M. Kosterlitz, *Phys. Rev. Lett.* 41, 128 (1978).
- (3) T. Niemeijer and J. M. J. Leeuwen, in "Phase Transition and Critical Phenomena", edited by C. Domb and M. S. Green, Academic, New York, Vol. 6, 425, 1976.
- (4) L. P. Kadanoff and A. Houghton, *Phys. Rev.* B11, 377 (1975).
- (5) Cheol Ju Kim, Dong J. Lee and Jong Myung Lee, To be published.
- (6) T. Schneider, Phys. Rev. B9. 381 (1974).