Phase Transitions in Lattice-Gas Models Far from Equilibrium

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A lattice-gas model with particle-conserving hopping dynamics on a periodic lattice is exposed to a strong external field along one of the principal axes. The resulting stationary state is determined exactly in the limit of infinite ratio of jump rates in and perpendicular to the field direction. In this state the gas-liquid phase transition is of mean-field type. For not too strong coupling in the direction orthogonal to the field, several other phase transitions occur. All of these have mean-field character as well.

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Recently Katz, Lebowitz, and Spohn[1] studied the gas-liquid phase transition of a lattice-gas model in a stationary nonequilibrium state. This state is produced by a constant uniform field along one of the principal directions of a periodic square lattice, which causes the flow of a steady current. Monte Carlo simulations show that for attractive interactions between the particles the critical temperature is raised above its equilibrium value, whereas for repulsive interactions it is lowered. A theoretical description of the phase transition in the nonequilibrium state has been thus far lacking.

Here we describe a slightly different version of this model, for which the stationary states can be found exactly. The gas-liquid phase transition turns out to be of mean-field type, as seems to be typical for nonequilibrium phase transitions.[2] It is our feeling that this model can serve as a prototype for phase transitions modified by the presence of stationary currents and as a testing ground for general theories describing such phase transitions. The model is described by configurations $\{\eta\}=\{\eta_{11}, \ldots, \eta_{NM}\}$, with η_{ij} either 0 or 1, defined on an $N\times M$ square lattice with periodic boundary conditions. The energy in equilibrium is given by

$$E(\{\eta\}) = \sum_{i=1}^{N}\sum_{j=1}^{M}(-4J_h\eta_{ij}\eta_{ij+1}-4J_v\eta_{ij}\eta_{i+1j}), \quad (1)$$

with $\eta_{N+1j}=\eta_{ij}$ and $\eta_{iM+1}=\eta_{i1}$. As in Ref. 1 we define a stochastic dynamics by allowing the particles to jump to unoccupied neighboring sites. In addition we restrict ourselves to the case that an infinite field is turned on in the vertical direction. Then the jump rates are defined as follows: The rate for a jump from (k,j) to $(i+1,j)$ is $\Gamma_{\|}$, that for a jump in the opposite direction vanishes, and the rates for jumps in the horizontal direction are given as

$$\Gamma(\{\eta\}\rightarrow\{\eta'\}) = \Gamma_{\perp}\exp\tfrac{1}{2}\beta[E(\{\eta\})-E(\{\eta'\})], \quad (2)$$

where η and η' must be connected through a jump from a site (i,j) to $(i,j\pm1)$. Note that these jump rates differ from the ones adopted in Ref. 1, but do satisfy the local detailed balance conditions imposed there. Next we pass to the limit $\Gamma_{\|}/\Gamma_{\perp}\rightarrow\infty$, corresponding to a strong enhancement of the jump rate in the field direction.[3] In this limit the distribution of particles in each single column relaxes on the time scale $\Gamma_{\|}^{-1}$ to the stationary distribution determined by the vertical jump process and the number of particles, say n, in the column. (Note that the vertical jump process is completely uninfluenced by the contents of the other columns.) This distribution simply gives equal weight to all configurations with n particles, since the numbers of configurations from which a given configuration can be gained and to which it can be lost are always equal to each other. (To be specific: both equal the number of clusters in the given configuration.)

On the time scale $\Gamma_{\perp}^{-1}$ the stationary distribution of the columns is disturbed by horizontal jumps. Between two consecutive jumps involving the same column, however, the column returns rapidly to a stationary condition; hence the horizontal jumps can be considered to occur among columns in a stationary single-column state. Hence on this time scale microscopic states of the full lattice can be specified by the occupation numbers n_j of all the columns.

For the probabilities $P(\{n\},t)\equiv P(n_1,\ldots,n_M,t)$ to find, at time t, n_j particles in column j, with

$j=1,\ldots,M$, there is a master equation of the form

$$\frac{\partial}{\partial t}P(\{n\},t)=\sum_{\{n'\}}[\Gamma(\{n'\}\rightarrow\{n\})P(\{n'\},t)-\Gamma(\{n\}\rightarrow\{n'\})P(\{n\},t)]. \tag{3}$$

To calculate the jump rates, consider a situation where the particle numbers in four consecutive columns are p, n, m, and q, respectively. Then the rate for a jump from the second to the third column is

$$\begin{aligned}\Gamma_{pq}(n,m\rightarrow n-1,m+1)=\Gamma_{\perp}n\frac{(N-m)}{N}\left\{\frac{q}{N}x+\frac{(N-q)}{N}x^{-1}\right\}\left\{\frac{P}{N}x^{-1}+\frac{N-p}{N}x\right\}\\ \times\left\{\frac{m(m-1)}{(N-1)(N-2)}y^{2}+\frac{2m(N-m-1)}{(N-1)(N-2)}+\frac{(N-m-1)(N-m-2)}{(N-1)(N-2)}y^{-2}\right\}\\ \times\left\{\frac{(n-1)(n-2)}{(N-1)(N-2)}y^{-2}+\frac{2(n-1)(N-n)}{(N-1)(N-2)}+\frac{(N-n)(N-n-1)}{(N-1)(N-2)}y^{2}\right\},\end{aligned} \tag{4}$$

with $x=\exp(\beta J_h)$ and $y=\exp(\beta J_v)$. In this equation the factor n appears because each particle in the second column may jump. The factor $(N-m)/N$ results from the restriction that the site to which the jump occurs must be empty. Then, for example, the event in which the jumping particle adjoins an empty site in the first column and two occupied sites in the second column, while the receiving site neighbors two empty sites in the third column and an occupied one in the fourth column, has a probability

$$\frac{(N-p)}{N}\frac{(n-1)(n-2)}{(N-1)(N-2)}\frac{(N-m-1)(N-m-2)}{(N-1)(N-2)}\frac{q}{N}.$$

According to (2) the jump rate in this configuration equals $\Gamma_{\perp}\exp[\beta(2J_h-4J_v)]$. Collecting the contributions from all different local configurations at the instant of the jump one recovers (4). Notice the particle-hole symmetry of (4): The Γ's are invariant under the transformation $n_i\rightarrow N-n_i$. Stationary solutions to master equations generally are hard to find, unless they satisfy a detailed balance condition. Hence one may try and look for stationary solutions to (3) by satisfying the detailed balance condition $P_0(\{n\})/P_0(\{n'\})=\Gamma(\{n'\}\rightarrow\{n\})/\Gamma(\{n\}\rightarrow\{n'\})$. This can be done exactly in the case $J_h=0$ only. If one assumes the system to be weakly coupled to a particle reservoir, the stationary solution for this case assumes the form

$$\begin{aligned}P_0(\{n\}|J_h=0)=\frac{1}{Z}\prod_{i=1}^{M}\binom{N}{n_i}\exp[\beta\mu(n_i-\tfrac{1}{2}N)]\\ \times\prod_{k=0}^{n_i}\frac{(k-1)(k-2)y^{2}+2(k-1)(N-k)+(N-k)(N-k-1)y^{-2}}{(k-1)(k-2)y^{-2}+2(k-1)(N-k)+(N-k)(N-k-1)y^{2}},\end{aligned} \tag{5}$$

where μ is a chemical potential and Z a normalization constant. Notice that (5) factorizes into independent distributions for the single columns. For $J_h\neq 0$ one has to pass to the limit $N\rightarrow\infty$ to be able to satisfy the detailed balance condition. Then, in a homogeneous phase the occupation number of each column shows only minor relative fluctuations about the average occupation number, say $\bar{n}$. Expanding the logarithm of $\Gamma(\{n\}\rightarrow\{n'\})$ to first order in deviations of the n_j around $\bar{n}$, one obtains the result

$$\frac{\Gamma_{pq}(n,m\rightarrow n-1,m+1)}{\Gamma_{pq}(n-1,m+1\rightarrow n,m)}\cong\frac{n}{N-n+1}\frac{N-m}{m+1}\exp\left[\frac{-\{4h(p-q)+8v(n-m)\}}{N}\right], \tag{6}$$

with $h=K_h(1-\phi^2K_h^2)^{-1}$ and $v=K_v(1-\phi^2k_v^2)^{-1}$, where $K_h=\tanh\beta J_h$; $K_v=\tanh\beta j_v$ and $\phi=(\bar{n}-N/2)/(N/2)$. Under this approximation the detailed balance condition is satisfied by the stationary distribution

$$P_0(\{n\})=\frac{1}{Z}\prod_{i=1}^{M}\binom{N}{n_i}e^{\beta\tilde{\mu}\Delta n_i}\exp(\{4h[\tfrac{1}{2}(\Delta n_i)^2+\Delta n_i\Delta n_{i+1}]+4v(\Delta n_i)^2\}/N) \tag{7}$$

with $\Delta n_i=n_i-\bar{n}$. From this equation the average occupation number $\bar{n}$ can be determined self-consistently as a function of $\tilde{\mu}$. Hence, $\bar{n}$, h, and v depend on $\tilde{\mu}$ which, therefore, cannot be identified with the chemical potential.

Since the field stirs the columns so effectively and thereby suppresses nearest-neighbor correlations almost completely, one may expect the gas-liquid phase transition to be of mean-field type. This is confirmed by explicit calculations. For determining the critical temperature we transform to discrete density waves $\hat{n}_k = \sum_{j=1}^{M} e^{-ikj}\Delta n_j$, with $k = 2\pi m/N$ and, for even M, m is chosen from the values $-\frac{1}{2}M+1, \ldots -1, 0, +\ldots, \frac{1}{2}M$. The stationary distribution can be expressed in terms of these variables as

$$P_0(\{\hat{n}_k\}) = \frac{1}{Z}\left(\frac{N}{\bar{n}}\right)^M \exp\left[-\sum_k \frac{\alpha_k}{M}\hat{n}_k \bar{n}_{-k}\right] \tag{8}$$

where $\alpha_k = 2\{1/(1-\phi^2) - h(1+2\cos k) - 2v\}/N$. The logarithms of the binomial factors were also expanded to first order in $n_j - \bar{n}$ and Stirling's approximation was used. Further, $\tilde{\mu}$ was identified as $\tilde{\mu} = \beta^{-1}\ln\{(1+\phi)/(1-\phi)\}$. In view of the particle-hole symmetry the critical point has to occur for $\tilde{\mu} = 0$, implying $\phi = 0$. The critical temperature can be determined from the condition that either, for $J_h \geq 0$, the compressibility $\langle \hat{n}_0^2 \rangle$ (with the brackets indicating an average over P_0), or, for $J_h \leq 0$, the "staggered compressibility" $\langle \hat{n}_\pi^2 \rangle$ goes to infinity. As one sees from (8) this leads to the conditions

$$2K_v^c + 3K_h^c = 1 \quad (J_h \geq 0), \tag{9a}$$

$$2K_v^c - K_h^c = 1 \quad (J_h \leq 0). \tag{9b}$$

In the isotropic attractive case, for example, this yields a critical temperature $T_c \cong 0.488J/k_B$, with k_B Boltzmann's constant, as compared to the equilibrium temperature $T_c^{eq} \cong 0.227J/k_B$. In contrast to the equilibrium case there is a phase transition at nonzero critical temperature also for $J_h = 0$ and $J_v > 0$ as well as for $J_v = 0$ and $J_h > 0$. It is also noteworthy that one need not take the limit $M \to \infty$ to obtain the phase transition; it occurs already for $M = 2$. This is consistent with the mean-field character of the transition.

In the following calculations we restrict ourselves to the case $J_h \geq 0$, noting, however, that the case $J_h \leq 0$ can be treated in a completely analogous way. The coexistence curve $\phi_{co}(T)$, with $T < T_c$, is obtained as usual with the aid of the Maxwell construction. Using particle-hole symmetry one sets $\mu(\phi_{co}) = \mu(-\phi_{co}) = \mu(0) = 0$, with μ the real chemical potential, different from $\tilde{\mu}$, and employs the compressibility relation $\partial\beta\mu/\partial\phi = \alpha_0$ to calculate $\mu(\phi_{co})$. This leads to a quadratic equation for ϕ_{co}^2, which close to the critical point reduces to

$$\phi_{co}^2 = -3\{2(\partial K_v/\partial T) + 3(\partial K_h/\partial T)\}_{T=T_c}(1 - 2K_v^3 - 3K_h^3)^{-1}_{T=T_c}(T_c - T). \tag{10}$$

From this one sees immediately that the critical exponent β, defined by $\phi_{co} \sim (T_c - T)^\beta$ as $T \to T_c$, has the mean-field value $\frac{1}{2}$.[4] The compressibility in the neighborhood of the critical point also is of typical mean-field form, i.e., $\kappa_T = MN/(2\alpha_0) = A^{\pm}|T - T_c|^{-1}$, with $A^+ = -\{8(\partial K_v/\partial T) + 12(\partial K_h/\partial T)\}^{-1}_{T=T_c}$ for $T > T_c$ on the critical isochore, and $A^- = 2A^+$ for $T < T_c$ on the coexistence curve. The specific heat, defined as the temperature derivative of the internal energy (1), exhibits the usual jump from the constant-value zero for $T > T_c$ to $-NM\partial/\partial T\{(J_h + J_v)\phi_{co}^2\}$ for $T < T_c$.

The stationary current is found from $J = \sum_{j=1}^{M} \Gamma_{\parallel}\langle \eta_{ij}(1 - \eta_{i+1\,j})\rangle$, which becomes

$$J = \tfrac{1}{4}\Gamma_{\parallel} M(1 - \phi^2) + O(MN^{-1/2}). \tag{11}$$

Hence, for a half-filled lattice the slope of J as a function of temperature shows a jump at $T = T_c$ and in general such a jump occurs as soon as one enters the coexistence region. The correlation function between column occupation numbers can be obtained easily from (7), e.g., with the aid of transfer-matrix methods. One finds the result

$$\langle (n_i - \bar{n})(n_j - \bar{n})\rangle = \exp(-|i-j|\kappa)\tanh(\kappa/2)M/2\alpha_0 \tag{12}$$

with $\kappa = \ln[\{(8h + \alpha_0)^{1/2} - \alpha_0^{1/2}\}/\{(8h + \alpha_0)^{1/2} + \alpha_0^{1/2}\}]$. Thus the correlation length $1/\kappa$ diverges as $|T - T_c|^{-1/2}$ near the critical point, in agreement again with mean-field theory. Furthermore, the fluctuation in the occupation number of a single column, obtained by setting $i = j$ in (12), also diverges as $|T - T_c|^{-1/2}$ near the critical temperature. As a consequence the correction term in (11) exhibits a similar divergence near T_c, which may be important for computer simulations on finite lattices.

Generalization of our model to d-dimensional simple cubic lattices is straightforward, provided the horizontal coupling constants in all the principal directions orthogonal to the field are equal. Then the critical temperature is determined by the conditions $2K_v + (2d-1)K_h = 1$, for $K_h > 0$, or $2K_v - (2d-3)K_h = 1$, for $k_h < 0$. This means that the

absolute value of T_c increases with dimensionality, but the enhancement relative to the equilibrium critical temperature decreases.

An intriguing feature of the phase transition studied here is that, despite its simple mean-field character, it has a very rich phase structure. First note that for $J_h = 0$ below T_c there are infinitely many pure states, as each of the columns orders independently into either the gaseous or the liquid phase. This degeneracy is not removed as soon as J_h is given a small nonzero value. For example, for positive J_h the staggered state remains stable for all temperatures below a critical temperature, given by (9b) for $J_h > 0$ as well. Stable means really stable, and not metastable, because to bring any column from the gaseous into the liquid phase or vice versa, one has to pass a barrier of infinite height. Further, just as for $J_h = 0$, there is an infinity of other stable states, each with its own critical temperature, which can be characterized by specifying for each column whether it is in the gaseous or in the liquid phase.

Other interesting questions are connected with the dynamics of system. One may ask for its dynamical critical behavior in case of a conserved order parameter (uniform ordered state) or a nonconserved one (staggered ordered state). Questions of stability and metastability also are of obvious interest. The boundary between metastability and instability, the *spinodal,* can be determined from the conditions $\alpha_0 = 0$ $(J_h > 0)$ or $\alpha_\pi = 0$ $(J_h < 0)$. Especially intriguing is what will happen to a less-favored stable state (e.g., the staggered state in case $J_h > 0$) when temperature is raised through its critical temperature.

To be noted further is the point that the value of the critical temperature depends strongly on the choice of the jump rates. Thus for the rates chosen in (1) there is no phase transition for $J_h = 0$.

Finally, there remain several problems that cannot be answered just by considering our simple model. The most obvious one, perhaps, is the question where and how the critical behavior crosses over from Ising-type to mean-field type behavior as the strength of the field is increased. According to Onuki and Kawasaki[2] the introduction of a uniform shearing, however small, immediately changes the character of a gas-liquid phase transition to mean-field type, but whether the introduction of a small field along one of the principles axes in the lattice gas has a similar effect remains to be proved.

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[1]S. Katz, J. L. Lebowitz, and H. Spohn, Phys. Rev. B **28**, 1655 (1983), and J. Stat. Phys. **34**, 497 (1984).

[2]A. Onuki and K. Kawasaki, Ann. Phys. (N.Y.) **121**, 456 (1979); K. Kawasaki and A. Onuki, in *Dynamical Critical Phenomena and Related Topics,* edited by Charles P. Enz, Lecture Notes in Physics, edited by J. Ehlers, Vol. 104 (Springer-Verlag, Berlin, 1979).

[3]For the considerations of this paper it is important that this limit is taken before the thermodynamic limit $N \to \infty$. It would be of interest to know what happens when these limits are interchanged.

[4]H. E. Stanley, *Introduction to Phase Transitions and Critical Phenomena* (Oxford Univ. Press, London, 1971).