Renormalization-group theory of spinodal decomposition

Gene F. Mazenko

The James Franck Institute and The Department of Physics, The University of Chicago, Chicago, Illinois 60637

Oriol T. Valls

School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455* and Argonne National Laboratory, Argonne, Illinois 60479

F. C. Zhang[†]

School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455 (Received 8 November 1984)

Renormalization-group (RG) methods developed previously for the study of the growth of order in unstable systems are extended to treat the spinodal decomposition of the two-dimensional spinexchange kinetic Ising model. The conservation of the order parameter and fixed-length sum rule are properly preserved in the theory. Various correlation functions in both coordinate and momentum space are calculated as functions of time. The scaling function for the structure factor is extracted. We compare our results with direct Monte Carlo (MC) simulations and find them in good agreement. The time rescaling parameter entering the RG analysis is temperature dependent, as was determined in previous work through a RG analysis of MC simulations. The results exhibit a longtime logarithmic growth law for the typical domain size, both analytically and numerically. In the time region where MC simulations have previously been performed, the logarithmic growth law can be fitted to a power law with an effective exponent. This exponent is found to be in excellent agreement with the result of MC simulations. The logarithmic growth law agrees with a physical model of interfacial motion which involves an interplay between the local curvature and an activated jump across the interface.

I. INTRODUCTION

The purpose of this paper is to study the spinodal decomposition of a binary alloy at 50% composition. This problem has been the object of considerable research effort (as reviewed, for example, in Refs. 1–13). The kinetic Ising model with spin-conserving dynamics has been extensively studied as a simple representation of the binary alloy problem. Results can be compared with Monte Carlo (MC) simulations which can be performed for this model in a straightforward manner. Our objective in this paper is to calculate observable quantities such as the quasistatic structure factor and several short-range spatial correlation functions for an Ising system on a square lattice subjected to a sudden quench from infinite temperature to some final temperature $T_F < T_c$ (T_c is the critical temperature), with a conserved order parameter.

In a recent paper⁴ we have carried out a renormalization-group (RG) analysis of MC simulations for a two-dimensional kinetic Ising model with both conserved (COP) and nonconserved (NCOP) order parameter. We demonstrated how the long-time growth laws in the scaling regime can be extracted from such an analysis. For the case of spinodal decomposition (COP) we found that the characteristic length L(t), or typical domain size, grows logarithmically with time. We will show here, how to calculate the correlation functions for the same conserving model as in Ref. 4 as a function of time. We will see that the position $q_m(t) \sim L^{-1}(t)$ of the peak in the

quasistatic structure factor $C(\mathbf{q},t)$ decreases with time according to a logarithmic law. However, this behavior becomes evident only over several decades in time. Over the time range in which MC simulations for the system under consideration have been performed,⁵ we find that an excellent fit to a power law is possible with an apparent exponent a=0.19. Thus, we conclude that MC simulations have not been performed over times long enough for the logarithmic behavior to be evident.

We will use the RG recursion relation approach developed⁶⁻¹⁰ previously for the study of growth kinetics. The perturbative scheme developed in Refs. 6 and 7 for spin-flip (SF) dynamics (a NCOP) cannot be used in the case of spinodal decomposition, where one has a COP, because the breaking up of the system into cells would introduce artificial in-cell conservation laws. It is desirable, however, to maintain the basic structure of the theory developed in Refs. 6 and 7. It was found there that the structure factor satisfies a recursion relation whose solution exhibits the main qualitative features for the case of a NCOP: (i) Development of a peak centered about the ordering wave number which grows in height and narrows with increasing time. (ii) Scaling behavior associated with the peak. (iii) Equilibration of the wave-number components away from the peak. (iv) Identification of the peak with the building of a Bragg peak characteristic of the new order in the system. The theory was reformulated in a form independent of the perturbation-theory approach in a qualitative fashion in Ref. 8 within the context of a COP. A more detailed and rigorous approach to this question was developed (for a NCOP) in Refs. 9 and 10. In Ref. 10 we considered an order-disorder transition model, still with a NCOP but with a conservation law included. The methods presented in Ref. 10 will be applied here, after straightforward modifications, to spinodal decomposition.

An important problem in the derivation of the recursion relation for the structure factor is the determination of the time rescaling factor $\Delta(b)$, which establishes how time must be rescaled when space is rescaled by a factor band self-similar behavior is to be obtained. Δ depends on the nature of the dynamics considered, of course. Because the perturbation-theory analysis of Refs. 6 and 7 is very local in nature, it cannot distinguish (as discussed in Refs. 8 and 9 between the factor Δ associated with growth kinetics far from equilibrium and the usual rescaling factor associated with fluctuations in equilibrium, which are typically not the same. For the case of a COP the question of the value of Δ was left open in Ref. 8. For a NCOP a theoretical argument backed by MC simulations was given in Ref. 9, showing that $\Delta = b^{-2}$, in two dimensions, which leads to the curvature-driven Lifshitz-Cahn-Allen¹¹ (LCA) growth law $L(t) \sim t^{1/2}$. The same determination was used in Ref. 10, where we studied the antiferromagnetic-spin-exchange (AFSE) kinetic Ising model, which still has a NCOP and exhibits the LCA growth law. The results of Refs. 9 and 10 were in good agreement with those of MC simulations.

For the case of ferromagnetic-spin-exchange (FSE) dynamics,¹² which is needed to model spinodal decomposition, the simple arguments used in Refs. 9 and 10 to obtain Δ are insufficient. For this reason we developed in Ref. 4 a new procedure for determining Δ , based on a RG analysis of MC simulations. For the NCOP case we were led, as expected, to the same results as obtained in Refs. 9 and 10, while for the COP case a very different result was obtained, leading to logarithmic growth laws. We will use Δ as determined in Ref. 4 in this paper. At the same time we will relate the nonperturbative approach to the recursion relations (Refs. 8-10) to the RG ideas of Ref. 4, thereby eliminating any logical gap which may have been present. Thus, we will show that the factor Δ found in Ref. 4 is indeed the same quantity introduced in the recursion relations used in previous work, and in this paper.

We show in Sec. IV of this paper that the logarithmic law can also be heuristically understood from a model for interfacial motion where diffusion across the interface is activated, and driven by the local curvature. As we pointed out in Ref. 4, this rather unexpected result is not necessarily in contradiction with the many published experimental and MC results reporting power-law growth in time $L(t) \sim t^a$. A review of the literature⁴ fails to show evidence for any definite value of a, and we concluded in Ref. 4 that the exponents reported represented the approximate result which one obtained when attempting to fit a logarithmic law to a power law over a restricted time range. One of the key results of the present paper is the verification of this conclusion through a direct calculation of the structure factor, as explained above.

This paper is organized as follows: In Sec. II we first

discuss the model and dynamics we use and the connection between our RG methods and textbook formulations of the RG. We then develop the recursion relations satisfied by $C(\mathbf{q},t)$ and several short-range correlation functions. Particular attention is given to connecting the work in Ref. 4 with that in Refs. 8-10. In Sec. III we discuss some of the analytic consequences of the recursion relations and then proceed to present the numerical results for the correlation functions. We show that our results compare quite well with those of available MC simulations⁵ in the appropriate time range. The question of apparent power-law behavior versus ultimate logarithmic growth is discussed in detail, and effective exponents in agreement with Ref. 5 are obtained. A brief conclusions section summarizes our results and gives a physical model of the interface which leads to the logarithmic growth resulting from the RG analysis.

II. RECURSION RELATIONS AND RENORMALIZATION-GROUP THEORY

We consider the standard¹³ lattice-gas representation of a binary alloy where a variable $\sigma(\mathbf{n})$ at site \mathbf{n} will be +1, or -1, if the site is occupied by an A or B atom. We assume a square lattice with unit lattice constant and nearest-neighbor interactions only, with coupling constant $K = -\beta J$, where $\beta = 1/k_B T$ and J is the value of the interaction for two atoms separated by a lattice spacing. We assume 50% concentration (which corresponds to zero external magnetic field in the associated spin problem). Phase separation (spinodal decomposition) takes place for a positive or ferromagnetic (K > 0) coupling. The system is driven by Kawasaki or FSE dynamics.¹² For the case of very fast quenches to a final temperature T_F (i.e., coupling K_F) the nearest-neighbor exchange probability represents the interaction of the system with a thermal bath at temperature T_F . We take the exchange probability per unit time between two nearest neighbors at sites n and **m** to be of the form¹²

$$W_{m,n}(\sigma) = \frac{1}{2} [1 - \tanh(\Delta E/2)],$$
 (2.1)

where ΔE is the change in energy associated with the exchange of the two spins, **m** and **n**, and is proportional to K_F . Note that (2.1) defines our unit of time.¹³ This dynamics conserves the order parameter (the total magnetization, or the number of particles of a given type). It therefore represents spinodal decomposition.

We will examine quenches from a perfectly disordered state (K=0) to a final value $K_F > K_c$, where $\tanh K_0 = (\sqrt{2}-1)$ is the value of the critical coupling. The quantity we will focus on is the quasistatic structure factor

$$C(\mathbf{q},t) = \frac{1}{N^2} \sum_{\mathbf{m},\mathbf{n}} e^{i\mathbf{q}\cdot(\mathbf{m}-\mathbf{m})} \epsilon(\mathbf{m}-\mathbf{n},t) , \qquad (2.2)$$

where

$$\boldsymbol{\epsilon}(\mathbf{m}-\mathbf{n},t) = \langle \sigma(\mathbf{m})\sigma(\mathbf{n}) \rangle_t . \qquad (2.3)$$

 N^2 is the number of sites and $\langle \rangle_t$ the average over the time-dependent probability distribution. This quantity is

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accessible experimentally by means of x-ray diffraction and has also been studied by MC simulations⁵ for precisely the model we consider here. We also consider shortrange correlations in coordinate space. This model with antiferromagnetic (rather than ferromagnetic) coupling corresponds¹⁰ to an order-disorder transition in a binary alloy.

Our objective here is to write a recursion relation for $C(\mathbf{q},t)$, that is an equation relating¹⁵ $C(\mathbf{q},t)$ to $C(\mathbf{q}',t')$, where q' and t' are rescaled wave vectors and times. It is important to establish this recursion relation from a nonperturbative point of view, particularly as it applies to spinodal decomposition, since only then can the conservation law be enforced in a global manner. Perturbation-theoryindependent renormalization-group methods were presented in detail in Sec. II of Ref. 9. In particular, the determination of the renormalized couplings K'(K) and of equilibrium quantities is discussed there and we will not repeat it. We will now, however, discuss the relationship between our nonperturbative RG analysis and standard field-theory RG methods¹⁶ of the field-theoretic type. This will enable us to make the connection between our procedure and standard RG theory.

To clarify this question, let us take, for simplicity, the case of a quench to zero temperature and a nonconserved order parameter. As in Ref. 4, let us introduce a quantity $R_M(t)$:¹⁷

$$R_{M}(t) = M^{-2d} \int_{0}^{M} d^{d}r_{2} \int_{0}^{M} d^{d}r_{1} [\epsilon(\mathbf{r}_{1} - \mathbf{r}_{2}, t) - \epsilon(\mathbf{r}_{1} - \mathbf{r}_{2}, 0)] . \quad (2.4)$$

In Eq. (2.4) *M* characterizes a length associated with some block of spins and *t* is the time after a quench. We then carry out a simple reparametrization and define

$$M' = M/b , \qquad (2.5a)$$

$$t' = t/b^{1/x}$$
, (2.5b)

where b > 0 and the exponent x is not yet specified. Since M = bM' and $t = b^{1/x}t'$, we have quite trivially

$$R_{M}(t) = R_{M'}(t',b)$$
 (2.6)

Equation (2.6) is formally similar to (8.1) in Ref. 16, which relates the renormalized $\Gamma_R^{(N)}$ and bare $\Gamma^{(N)}$ amputated *N*-point vertices (see Ref. 16 for notation and details) by an analogous reparametrization of scales.

Since the left-hand side of (2.6) is independent of b, the derivative of (2.6) with respect to b is zero, and, using the chain rule and (2.5), we obtain

$$-M'\frac{\partial}{\partial M'}R_{M'}(t',b) - \frac{t'}{x}\frac{\partial}{\partial t'}R_{M'}(t',b) + b\frac{\partial}{\partial b}R_{M'}(t',b) = 0. \quad (2.7)$$

If one can find a value of x such that, for large M and t, the last term on the left-hand side of (2.7) vanishes, then one has

$$-M'\frac{\partial}{\partial M'}R_{M'}(t',b)-\frac{t'}{x}\frac{\partial}{\partial t'}R_{M'}(t',b)=0, \qquad (2.8)$$

which has a solution of the form

$$R_{M'}(t',b) = f(M'/L(t')), \qquad (2.9)$$

where L(t') satisfies

$$x = \frac{\partial \ln L(t')}{\partial \ln t'} .$$
 (2.10)

This, in turn, has a solution

$$L(t') = L_0(t')^x . (2.11)$$

Alternatively, if there is scaling in the system and x is properly adjusted, (2.9) and (2.11) hold and one is driven to the fixed point

$$b\frac{\partial}{\partial b}R_{M'}(t',b) = 0 \tag{2.12}$$

in the limit of large t and M. The dropping of the last term on the left-hand side (lhs) of (2.7) is similar to the argument in treating the Callan-Symanzik equations (Ref. 16, p. 212), where, due to Weinberg's theorem, one can neglect the lhs of (8.105) in the scaling region $k_i/m \to \infty$. The scaling region in our case corresponds to large M and t.

The above analysis for the nonequilibrium case is well posed but difficult to implement since one needs to find an x such that (2.12) holds for large M and t. The nontrivial point here is to see that there is a dominant length L(t). In the field-theoretical version of the RG it is given that there is an infinite length ξ associated with a vanishing renormalized mass.

Since the above development is difficult to implement in practice, we have developed a more sophisticated and practical RG approach. By construction, $R_M(t)$ and $R_{M'}(t)$ are monotonic functions ranging from 0 to 1 as t evolves. Since M' < M, we will have $R_{M'}(t) > R_M(t)$. We can *define* a quantity t' such that

$$R_{M}(t) = R_{M'}(t') , \qquad (2.13)$$

where, in general,

$$t' = t'(M, b, t)$$
 (2.14)

Just as above, we can write an RG equation:

$$\frac{\partial}{\partial b}R_{M}(t) = 0 = -\frac{M'}{b}\frac{\partial}{\partial M'}R_{M'}(t') + \frac{\partial t'}{\partial b}\frac{\partial}{\partial t'}R_{M'}(t') .$$
(2.15)

This is a homogeneous RG equation of the same form as Eq. (8.11) in Ref. 16. Let us suppose that we can show that

$$-\frac{1}{x} = \frac{\partial \ln t'}{\partial \ln b}$$
(2.16)

is independent of M and t for large M and t. Then

$$\frac{\partial}{\partial \ln M'} R_{M'}(t') + \frac{1}{x_{\prime}} \frac{\partial}{\partial \ln t'} R_{M'}(t') = 0$$
(2.17)

again has a solution of the form (2.9). Since we expect $t' = \Delta(b)t$ for large t, we have from (2.16) that

$$-x^{-1} = \frac{\partial \ln \Delta(b)}{\partial \ln b} . \tag{2.18}$$

If x is independent of b, then

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$$\Delta(b) = b^{-1/x} , \qquad (2.19a)$$

$$t' = t/b^x$$
, (2.19b)

$$L(t') = L_0 t^{x} (b^{-x})^{x}, \qquad (2.19c)$$

and as $b \rightarrow 1$

$$L(t) = L_0 t^{\mathbf{x}} . \tag{2.20}$$

Therefore, showing that x as given by (2.16) is independent of M, for large M, is equivalent to showing that

$$b\frac{\partial}{\partial b}R(M',t',b) = 0 \qquad (2.21)$$

for large M and t, and gives both the scaling law and the exponent x, and identifies a growth kinetics fixed point.

We have actually implemented our RG analysis directly through the use of Eqs. (2.13) and (2.14), which can be conveniently studied using Monte Carlo simulations. Thus one can test whether $\Delta = t'(M,b,t)/t$ is independent of M for large M and t. It is straightforward to extend this analysis to include the effects of a quench to finite temperature as discussed in Ref. 4. There we verified that for sufficiently large M and t a time rescaling factor $\Delta(b,t)$ exists such that, with $t' = \Delta t$, one had

$$R_M(t) = \nu^2 R_{M/b}(t') , \qquad (2.22)$$

where $v = m_E(T_F)/m_E(T'_F)$ (Ref. 18) $[m_E(T_F)]$ is the spontaneous magnetization corresponding to the final equilibrium state]. Note that, if the temperature dependence is included, then $R_M(t)$ is a function of K_F , and $R_M(t')$ a function of K'_F (assuming $K_I = 0$).

For the quasistatic structure factor, the recursion relation was found to be of the form

$$C(\mathbf{q},t) = C_0(\mathbf{q},t) + P(\mathbf{q},t)C(bq,t') .$$
(2.23)

We will now discuss the origins of this equation, as it applies to spinodal decomposition, from a nonperturbative point of view. As in the case where the order parameter is not conserved (Refs. 9 and 10), we require that $C_0(\mathbf{q},t)$ and $P(\mathbf{q},t)$ be short ranged in time and space so that their spatial Fourier transforms $\epsilon_0(\mathbf{r},t)$ and $\pi(\mathbf{n},t)$ fall off rapidly with distance and approach their equilibrium values at coupling K_F , $\epsilon_0(\mathbf{n},K_F)$, on a short-time scale. The usefulness of (2.23) arises in those problems where growing structures are associated with some ordering wave number \mathbf{q}_0 , and $C(\mathbf{q}_0,t)$ becomes arbitrarily large for a sufficiently long time. We then have $C(\mathbf{q},t) \gg C_0(\mathbf{q},t)$ and $P(\mathbf{q},t) = P(\mathbf{q}_0,\infty) = P_F$ for \mathbf{q} near \mathbf{q}_0 , and (2.23) reduces to the scaling result

$$C(\mathbf{q},t) = P_F C(b\mathbf{q},t') . \tag{2.24}$$

In order to establish the form of (2.23) it is sufficient, from the theoretical point of view, to show that (2.24) is valid at long times and distances, since the form of (2.23) follows from (2.24) and the assumption of short-ranged ϵ_0 (\mathbf{r}, t) and $\pi(\mathbf{r}, t)$. We will now demonstrate that the validity of (2.24) follows from (2.22). In this demonstration we will, as a consequence, determine P_F and t'=t'(b,t).

How does one go from (2.22) to (2.24)? This can be accomplished by observing that (2.4) can be rewritten as [see

Eq. (2.6) in Ref. 4]

$$R_{M}(t) = \left[\prod_{i=1}^{d} \left[\frac{2}{M^{2}} \right] \int_{0}^{M} dr_{i}(M - r_{i}) \right] \times [\epsilon(\mathbf{r}_{1} - \mathbf{r}_{2}, t) - \epsilon(\mathbf{r}_{1} - \mathbf{r}_{2}, 0)], \qquad (2.25)$$

which can be generalized to a "rectangular" geometry:

$$R(\mathbf{M},t) = \left[\prod_{i=1}^{d} \frac{2}{M_i^2} \int_0^{M_i} dr_i (M_i - r_i) \right] \times [\boldsymbol{\epsilon}(\mathbf{r},t) - \boldsymbol{\epsilon}(\mathbf{r},0)] . \qquad (2.26)$$

It follows from (2.26) that

$$\epsilon(\mathbf{r},t) - \epsilon(\mathbf{r},0) = \frac{1}{2^d} \left[\prod_{i=1}^d \frac{\partial^2}{\partial r_i^2} r_i^2 \right] R(\mathbf{r},t) . \qquad (2.27)$$

Since for sufficiently large distances correlation functions are approximately isotropic, it is natural to assume that Eq. (2.22) can be generalized to

$$R_{\mathbf{M}}(t) = v^2 R_{\mathbf{M}/b}(t')$$
 (2.28)

From Eqs. (2.27) and (2.28) we obtain for large $|\mathbf{r}|$, using the fact that $\epsilon(\mathbf{r}, 0)$ vanishes rapidly for increasing $|\mathbf{r}|$,

$$\boldsymbol{\epsilon}(\mathbf{r},t) = v^2 \boldsymbol{\epsilon}(\mathbf{r}/b,t') . \tag{2.29}$$

Assuming that the large $|\mathbf{r}|$ part of $\epsilon(\mathbf{r},t)$ dominates the small **q** contributions to the structure factor in (2.2), we finally arrive at (2.24) with t'=t'(b,t) known and $P_F=b^d v^2$.

The scaling results (2.24), (2.28), and (2.29) can be used immediately to establish scaling forms, as, for example, in Refs. 9 and 10. Thus, the solution to (2.24) is of the form

$$C(\mathbf{q},t,\xi) = m_E^2 L^d(t,\xi) F(\mathbf{q}L(t,\xi),\xi/L(t,\xi)) , \qquad (2.30)$$

where F is a scaling function, and

$$L(t,\xi) = bL(t',\xi')$$
 (2.31)

is a characteristic length and we have explicitly included the K_F dependence through the equilibrium correlation length ξ . Given the time rescaling factor Δ , one can also make statements about the long-time growth law for L. If, as for the order-disorder case, $\Delta = b^{-x}$, then the growth law is given by

$$L = t^{1/x} f(\xi/t^{1/x})$$
.

The scaling relations do not, however, determine the scaling functions F or f, or the range in which they are valid. We obtain a complete theory by extending (2.29) to a recursion relation valid for all times and distances and which reduces to (2.29) in the appropriate limit. It follows from the above discussion, and that in Ref. 4, that the recursion relation (2.24) or (2.29) cannot be directly extended to arbitrary \mathbf{q} or \mathbf{r} and time. It is obvious, for example, that Eq. (2.29) will break down at short enough times, since the time derivative of $\epsilon(\mathbf{r}, t)$ evaluated at t = 0 does not satisfy scaling. Thus, a recursion relation valid

at all length and time scales must have a form such as (2.23) where the functions $C_0(\mathbf{q},t)$ and $P(\mathbf{q},t)$ must be determined from short-time and distance considerations in addition to the asymptotic forms (2.24) or (2.29). We will now discuss the determination of C_0 and P for the case of FSE dynamics.

The recursion relation (2.23) must reproduce the initial and final equilibrium structure factors for t=0 and as $t\to\infty$. As discussed in Ref. 9, one can then tie down the equilibrium forms of C_0 and P using their short-ranged nature, knowledge of the short-ranged correlations $\epsilon(\mathbf{r}, T_F)$, and the $\mathbf{q}=0$ value of $C(\mathbf{q})$. One then obtains excellent approximations for the equilibrium structure factor. Next, one must determine the time dependence of C_0 and P which takes them from their initial to their final values. In the conserved case, $C(\mathbf{q}=0, t)$ is time independent and care must be taken to preserve this and the fixed-length spin sum rule:

$$\int \frac{d^2q}{(2\pi)^2} C(\mathbf{q}, t) = 1 .$$
 (2.32)

To do this, we will follow essentially the same procedure for satisfying the sum rule in the presence of a conservation law that we developed in Ref. 10, where there is a conservation law, but it is not satisfied by the order parameter.

We first assume that $P(\mathbf{q},t)$ can be written in the relaxational form:

$$P(\mathbf{q},t) = P_F(\mathbf{q}) + e^{-\lambda(\mathbf{q})t} [P_I(\mathbf{q}) - P_F(\mathbf{q})], \qquad (2.33)$$

where $\lambda(\mathbf{q})$ is assumed to be of the form

$$\lambda(\mathbf{q}) = \lambda(1 - g_1(\mathbf{q})) \tag{2.34}$$

with

$$g_1(\mathbf{q}) = \frac{1}{2} (\cos q_x + \cos q_y) , \qquad (2.35)$$

so that the conservation law $[\lambda(\mathbf{q}) \approx O(q^2)]$ and the lattice periodicity are preserved. The quantity λ is a function of K_F , which we determine below. For C_0 we write

$$C_{0}(\mathbf{q},t) = C_{0,F}(\mathbf{q}) + e^{-\lambda(\mathbf{q})t} \times [C_{0,F}(\mathbf{q}) - C_{0,F}(\mathbf{q})] - B(t)G(\mathbf{q}) .$$
(2.36)

In Eqs. (2.33) and (2.36) the indices I and F denote the corresponding quantities at thermal equilibrium with couplings K_I and K_F , respectively. These quantities are given in Ref. 9. The last term in Eq. (2.9) is needed to ensure that the sum rule (2.32) is satisfied. Since this term is not present in thermal equilibrium, we must have $B(0)=B(\infty)=0$. The conservation law requires G(q=0) to vanish. It turns out to be convenient¹⁹ to impose

$$\lim_{q \to 0} q^{-2} G(\mathbf{q}) = 0 , \qquad (2.37)$$

and we can take, without loss of generality, the normalization of $G(\mathbf{q})$ to be

$$\int \frac{d^2 q}{(2\pi)^2} G(\mathbf{q}) = 1 .$$
 (2.38)

The parameter $\tilde{\lambda}$ in Eq. (2.34) and the functions $G(\mathbf{q})$ and B(t) can be determined from two conditions: (i) that the recursion relation (2.23) preserves, at all times, the sum rule, and (ii) that the initial derivative of the recursion relation (2.23) gives the correct exact result for the initial derivative of $C(\mathbf{q}, t)$, which, for $K_I = 0$, is given by

$$\Gamma(\mathbf{q}) = \frac{d}{dt} C(\mathbf{q}, t) \bigg|_{t=0}$$

= $[1-g_1(\mathbf{q})][1+4q_1(\mathbf{q})]$
 $\times [\frac{5}{4} \tanh(2K) + \tanh(4K) + \frac{1}{4} \tanh(6K)].$

(2.39)

After a brief calculation similar to that given in Sec. II of Ref. 10, and with the notation used there, one then obtains

$$\widetilde{\lambda} = \lim_{q \to 0} \frac{\Gamma(\mathbf{q}) - \Delta \Gamma'(b\mathbf{q})}{[1 - g_1(\mathbf{q})](2r_F + s_F)}$$
(2.40)

$$G(\mathbf{q}) = \frac{2}{\widetilde{\lambda}r_F} \{ \widetilde{\lambda} [1 - g_1(\mathbf{q})] [2r_F g_1(\mathbf{q}) + s_F g_2(\mathbf{q})] - \Gamma(\mathbf{q}) + \Delta f(\mathbf{q}) \Gamma'(b\mathbf{q}) / 4 \} .$$
(2.41)

One can easily verify that $\tilde{\lambda}$ is always a positive quantity. These formulas are valid even when Δ depends on *t*, provided only that the initial derivative of $\Delta(t)t$ equals $\Delta(0)$. In (2.40) and (2.41),

$$g_2(\mathbf{q}) = \cos q_x \cos q_y \quad , \tag{2.42}$$

$$f(\mathbf{q}) = 1 + 2g_1(\mathbf{q}) + g_2(\mathbf{q}) . \tag{2.43}$$

Equation (2.38) can be used to find B(t). However, since it is rather awkward to determine B(t) from an integral constraint $C(\mathbf{q},t)$, we consider, instead, the Fourier transform of the recursion relation, Eq. (2.23). We have

$$\epsilon(\mathbf{n},t) = \epsilon_0(\mathbf{n},t) - B(t)G(\mathbf{n}) + \sum_m \pi_{\mathbf{n}-b\mathbf{m}}(t)\epsilon'(\mathbf{m},\Delta t) , \qquad (2.44)$$

where $\epsilon_0(\mathbf{n},t)$ is the Fourier transform of the relaxational part of $C_0(\mathbf{q},t)$ [the first two terms on the right-hand side of Eq. (2.36)], $G(\mathbf{n})$ the Fourier transform of $G(\mathbf{q})$, and $\pi_{\mathbf{m}-b\mathbf{m}}(t)$ the Fourier component of $P(\mathbf{q},t)$ corresponding to the site $\mathbf{n}=b\mathbf{m}$. These quantities can all be evaluated analytically. The resulting expressions are precisely equivalent to those given in Appendix A of Ref. 10, except that the argument of the modified Bessel function involved is now $+\lambda t/2$ rather than $-\lambda t/2$. The timedependent parts of $\epsilon_0(\mathbf{n},t)$ and $\pi_{\mathbf{n},\mathbf{m}}(t)$ vanish only as t^{-1} as $t \to \infty$.

The sum rule (2.32) can be easily enforced in coordinate space by setting $\epsilon(0,t) = \epsilon'(0,\Delta t) = 1$ at all times in (2.44). This is computationally more practical than the integral constraint (2.32).

Iteration of (2.44), truncated at a sufficiently large $|\mathbf{n}|$

so as to obtain convergence, yields results for B(t), and for the short-range correlation functions $\epsilon(\mathbf{n}, t)$. Thus, for example, $\epsilon(1,0;t)$ is the nearest-neighbor correlation functions, proportional to the energy of the system in the present case. Once B(t) is known numerically, the structure factor can be obtained by iterating (2.23), taking into account (2.33) and (2.36). The initial and final equilibrium quantities are taken to be as given in Ref. 9.

As emphasized in Ref. 4 and above, a key difference between the ferromagnetic and the antiferromagnetic cases (phase separation versus an order-disorder transition) is the behavior of the time rescaling factor Δ . In Refs. 4, 9, and 10 we indicated that Δ can be taken as a constant in both time and temperature in the NCOP case. A key ingredient in this assessment is that as one renormalizes the final temperature T_F iterates to zero (for $T_F < T_c$), and for sufficiently long times it is only the zero-temperature values of Δ which will influence the long-time growth. In the case of spinodal decomposition matters are more involved because $\lim_{T_F\to 0} \Delta(T_F, t\to \infty) = 0$. Therefore, we must include the time dependence of Δ so that at $T_F = 0$ one has t' < t, and one iterates under renormalization to earlier nonzero times as at finite temperature. The longtime growth in this problem is governed⁴ by the approach of $\Delta(T_F, t = \infty)$ to zero as $T_F \rightarrow 0$. We must therefore include both the temperature and time dependence of Δ . The result we obtained in Ref. 4, for a rescaling factor b=2, which should be valid for quenches well below the critical point, is

$$\Delta(t,y) = 0.41y + \frac{0.057}{1+t/5} , \qquad (2.45)$$

where y is the low-temperature variable, $y = e^{-4K_F}$. Note that the time scale used here differs, as explained in Ref. 10, from the MC scale by a factor of 4.

The slight time dependence of Δ in (2.45) implies that, upon iteration of the recursion relations, the right-hand side of (2.33) and the corresponding term of (2.36) are not strictly relaxational in time. This is not an important effect because the difference is very slight: the time dependence of Δ is only appreciable at early times where the initial derivatives of $C(\mathbf{q},t)$ determine C_0 and P. Note that the time dependence of Δ does not influence the initial value of the time derivative, since only the combination $t'=\Delta(y,t)t$ enters in the equations. This completes our discussion of the recursion relation and we will proceed with their solution in the next section.

III. RESULTS

From the recursion relations (2.27) and (2.4) we can obtain, by following standard⁷ iterative procedures, our results for the time evolution of the correlation functions.

A. Scaling results

First we consider the analytic consequences of the recursion relation given by (2.23). At long times and small (but nonzero) wave vectors, it follows from the asymptotic behavior of C_0 and P (which can be elucidated from well-known asymptotic properties of Bessel functions) that (2.4) reduces, as it should, to the form (2.24). Therefore,

$$C(\mathbf{q},t,\xi) = b^d v^2 C'(b\mathbf{q},\Delta_{\infty}(\xi)t,\xi/b) , \qquad (3.1)$$

where we have written explicitly the dependence on the final quenching temperature through the correlation length ξ for the final equilibrium temperature. In (3.1) Δ_{∞} is the long-time value of $\Delta(y,t)$ given approximately by 0.41y at b=2. As in the nonconserved case, (3.1) has a solution of the scaling form (2.30) which we will write as

$$C(\mathbf{q},t,\xi) = C_m(t,\xi)F(\mathbf{q}L,\xi/L) , \qquad (3.2)$$

where $L(t,\xi) = q_M^{-1}(t,\xi)$ [q_M is the value of q for which $C(\mathbf{q},t)$ has a maximum at time t] satisfies

$$L(t,\xi) = bL(t',\xi')$$
, (3.3)

while C_m is the value of the maximum chosen such that $F(1, \xi/L) = 1$ and which satisfies

$$C_{M}(t,\xi) = b^{d} v^{2} C_{M}(t',\xi') . \qquad (3.4)$$

As discussed in Ref. 4 [see below (2.27) there], (3.3) has a solution

$$L(t,\xi) = \xi C_0 \ln(t\xi^{\beta} / \Delta^{1/(b-1)})$$
(3.5)

when

$$\Delta = \Delta_0 y^{\alpha} = \Delta_0 e^{-2\alpha/\xi} \tag{3.6}$$

for small ξ . In (3.6) α and Δ_0 must be functions of b and, in (3.5), $\beta = \Delta_0 / \ln b$. Note that in the low-temperature limit, where $\xi \rightarrow 0$, $L(t,\xi)$ goes to a finite, nonzero value. Consequently, for $t < \infty$ and $\xi = 0$, q_m will be a constant, while for $0 < \xi < \infty$ and t sufficiently large,

$$q_m(t) = (\ln t)^{-1}$$
 (3.7)

Equation (3.4) for the peak height has a solution of the form

$$C_m(t,\xi) = m_E^2 L^d(t,\xi) f_m(t\xi^\beta / \Delta^{1/(b-1)}).$$
(3.8)

In writing this solution in d dimensions we should emphasize that we have direct evidence for the behavior of Δ given by (3.6), only for two dimensions. We note, as in the discussion of $L(t,\xi)$, that as $\xi \to 0$ we expect $C_m(t,\xi)$ to be finite. Since as $\xi \to 0$ the argument of f_m goes to ∞ , we require that $f_m(\infty)$ be finite. Alternatively, for fixed ξ as $t \to \infty$ we find

$$C_m(t,\xi) = m_E^2 L^d(t,\xi) f_m(\infty) , \qquad (3.9)$$

and the scaling relation

$$\lim_{t \to \infty} C_m(t,\xi) q_m^{-d}(t,\xi) = m_E^2 f_m(\infty)$$
(3.10)

follows.

B. Numerical results

We now discuss the results we obtain by numerically iterating the recursion relations derived in Sec. II. We will present results for the nearest-neighbor correlation function, for some other short-range real-space correlations, and for the quasistatic structure factor. The results will be compared with the MC simulations of Ref. 5. Our numerical results show the logarithmic time dependence at finite temperature, and the freezing behavior at zero temperature, which are expected from our theoretical analysis for the temperature dependence of the time rescaling parameter Δ . We will verify the existence of scaling behavior and extract the shape function for C(q,t).

All calculations have been performed with the spatial rescaling factor b set to b=2. Te time rescaling factor in this case is given by (2.45). We take $K_I=0$, and the values of K_F are indicated by $u_F=\tanh K_F$.

The first step is to use the matrix recursion relation (2.44) with the sum-rule condition imposed as explained in Sec. II to obtain the function B(t). This is done, in principle, as in Ref. 10. However, a larger number of equations must be kept in order to obtain a convergent result for B(t) because the time-dependent part of the quantities $\pi_{m,n}(t)$ in (2.44) decays only as t^{-1} at long times in the present case, rather than according to a t^{-3} law as they did in the order-disorder problem. Up to 13 short-range correlation functions must now be included in the calculation. This ensures accurate results for B(t) and the first few short-range correlation functions.

The resulting B(t) is shown in Fig. 1 for $u_F=1$ and $u_F=0.5$. Its maximum value is about 0.25 at $t \sim 0.5$, and it decreases fairly quickly as t increases further. Furthermore, B(t) is weakly temperature dependent.

In Fig. 2 we show the nearest-neighbor correlation function $\epsilon(1,0;t)$ (solid curve) for $u_F = 0.636$, which corresponds to $T_F = 0.59 T_c$, the case studied in Ref. 5. We have also plotted (dots) the MC results of Ref. 5 in the same time range (with MC steps converted to our units). The MC results of Ref. 5 were obtained on an 80×80 system, averaging over only eight runs. The agreement with our results is quite satisfactory, particularly at long times: Because of the effects of metastable configurations in fin-



FIG. 1. The function B(t), determined from the sum-rule condition, for two different values of $u_F = \tanh K_F$, as a function of time. The solid line is for $u_F = 0.5$, and the dashed line for $u_F = 1$.



FIG. 2. Our theoretical results for the nearest-neighbor correlation function (solid line) at $T_F=0.59T_c$ compared with the Monte Carlo results (dots) from Ref. 5 at the same temperature.

ite systems MC simulation results for $\epsilon(1,0;t)$ are known to be²⁰ below the correct values even in the order-disorder case, and this effect should be more troublesome for spinodal decomposition since the conservation law will produce, for a finite system, many additional possible metastable states.

The evolution of several other short-range correlation functions is plotted in Fig. 3. Because of the COP, they all evolve much more slowly than in the order-disorder problem. The scaling regime is not reached until fairly long times. The conservation law for C(q=0,t), which reads, in coordinate space,

$$\sum_{\mathbf{n}=0} \epsilon(\mathbf{n},t) = 0 , \qquad (3.11)$$

implies, since $\epsilon(1,0;t) > 0$, that several of the short-range correlation functions must be negative at very early times. This result is indeed verified (see inset of Fig. 3). As time increases, the shorter-ranged correlation functions become positive, while in compensation relatively longer-range ones become more negative. An analytic calculation shows that only the nearest-neighbor correlation function has a positive initial time derivative.



FIG. 3. Several short-range correlation functions at $u_F = 0.5$ from our theory. The inset is for early times.



FIG. 4. Theoretical results for the quasistatic structure factor, for $u_F = 0.5$ at several times.

We now turn to the quasistatic structure factor, which is the quantity we want to focus on. After having obtained numerical values for B(t) as explained above, the recursion relation (2.23) is readily solved by iteration. In Fig. 4 we show $C(\mathbf{q},t)$ as a function of $q_x = q_y$ and several times for a quench to $u_F=0.5$. Of course, at $\mathbf{q}=0$, $C(\mathbf{q},t)$ is independent of time because of the conservation law. This constant is unity at $u_I=0$ (which is the only case we consider here). The peak indicating the growth of order, therefore, cannot grow at $\mathbf{q}=0$ (as in the orderdisorder problem), but it grows at a finite value of qwhich decreases with time as the peak becomes sharper but never vanishes.

In Ref. 4 and in Sec. III A we have discussed the expected behavior of the peak position $q_m(t)$ at long times. We have extensively studied this time and temperature dependence of $q_m(t)$ and verified our analytic predictions.



FIG. 5. The peak position of the quasistatic structure factor versus $(\ln t)^{-1}$ in the time regime $t \sim 10^2$ to 10^{27} , as extracted from the numerical results for $C(\mathbf{q}, t)$.



FIG. 6. The peak position (lower dots) and the height (upper dots) of the quasielastic structure factor as functions of time (for $u_F = 0.5$) in the time region of the simulations in Ref. 5. The corresponding solid lines are a power-law fit [see Eq. (3.12)].

Thus, in Fig. 5 we plot $q_m(t)$ as a function of $(\ln t)^{-1}$ for a final temperature $T_F = 0.59 T_c$. The dots are the results for $C(\mathbf{q},t)$ as obtained from the recursion relation (2.4). A very extensive time region $(100 < t < 10^{27})$ is included in the plot. The straight line corresponds to proportionality between q_m and $(\ln t)^{-1}$. Except for the wiggles around this line, which are an artifact resulting from the use of an integer value (b=2) (Ref. 21) for the spatial rescaling factor, we see that the proportionality between the two quantities involved is well verified.

We must emphasize that such behavior is only clearly noticeable when one looks over many decades of time. Over shorter intervals, growth appears to be characterized by a power law. Thus our results are not, as we shall see, in contradiction with those of MC simulations, which have been performed only over a much narrower time range, $t \leq 10^4$ (in our units). This is shown in Fig. 6, which is a log-log plot of $q_m(t)$ versus t for this time range, at $u_F = 0.5$. The straight line is a power-law fit,

$$q_m(t) \propto t^{-a} , \qquad (3.12)$$

corresponding to an exponent a = 0.19. The fit appears to be excellent. We also plotted in Fig. 6 the value of $C_m(t) = C(q_m(t), t)$ and again observed an excellent power-law fit of the form $C_m \propto t^{a'}$ with a'=2a, which is required by scaling [see Eq. (3.10) and discussion below]. The same values are obtained in the same time range for quenching to the final temperature, $T_F = 0.59 T_c$, chosen in the MC simulations.⁵ The result a = 0.19 is in excellent agreement with the value a = 0.2 found in the MC simulation. The value of a' quoted in Ref. 5, however, is $a' \neq da$. We believe that this apparent discrepancy is due to the difficulty in estimating C_m and q_m in the fairly widely spaced wave-number mesh used in Ref. 5. It is also instructive to compare C(q,t), obtained here, with the corresponding MC results.⁵ This is done in Fig. 7: The solid line represents our results as a function of q_x/π (and $q_y = q_x$) at $T_F = 0.59T_c$ and time t = 24763, in our units. The dashed lines join the points corresponding to



FIG. 7. The theoretical result for the quasistatic structure factor for $T_F=0.59T_c$ and at t=24763 (solid line) compared with the Monte Carlo result (dashed line) from Ref. 5 for the same temperature and at the same time.

the results obtained in Ref. 5 for the same time and quenching temperature. The two results are, in our opinion, in good agreement. The MC results (which again represent an average over only eight runs) appear to indicate a narrower peak of about the same height, but the actual difference should be less since our result properly satisfies the sum rule (2.32). However, the main features of the curves (peak position and height) are in good agreement.

When we quench to zero temperature $(u_F = 1)$ we get, as expected, a completely different behavior which further verifies our analytic results. After relatively rapid growth at early $(t \le 60)$ times both $q_m(t)$ and $C_m(t)$ approach a constant value. We have tested this freezing up to $t=10^{30}$. Thus, the system will never reach equilibrium under Kawasaki dynamics when quenched to zero temperature.

All of these results are obtained using the recursion relation (2.4) and the result for the time rescaling parameter $\Delta(y,t)$ obtained from short-time MC simulations in Ref. 7. There are no adjustable parameters. The growth of order is governed only by the structure of the recursion relations. The agreement between theory and simulations, with respect to power-law exponents, $C(\mathbf{q},t)$, and $\epsilon(1,0;t)$ reflects the degree to which our method can quantitatively reproduce the simulation results. It is certainly desirable to have additional MC data available since there were only eight runs in Ref. 5 on a fairly coarse **q** mesh. Given the resulting statistical uncertainty (reflected in the apparent lack of scaling), we conclude that the agreement is as good as can be expected.

We turn now to the scaling properties of $C(\mathbf{q},t)$ for the case of spinodal decomposition. The asymptotic solution of the recursion relation is given by (3.10). This equation and the sum rule imply that $C_m(t)q_m^d(t)$ should be in-



FIG. 8. The scaling function F(x) [Eq. (3.2)] as a function of $|\mathbf{x}|$. The solid and dashed lines corresponds to $u_F=0.5$ and $u_F=0.64$, respectively. The dots are a fit of the form given by Eq. (3.13).

dependent of time (except at early times). We find that $C_m(t)q_m^2(t)$ is a constant for t > 100 over 25 decades in time. In particular, within any restricted time region where one observes apparent power-law behavior, we find a'=2a.

We have also extracted the scaling function F(x) $[x \equiv q/q_m(t)]$ defined by (3.2). In Fig. 8 we have plotted this function for two values of u_F ($u_F=0.5$ and 0.636). We can see that F(x) is only weakly temperature dependent. This was also found to hold in the order-disorder case, although, of course, the general shape of F(x) was very different.

For x > 1, i.e., $q > q_m$, the tail of the shape function can be fitted very well to the form

$$F(x) = \frac{1}{\alpha_1 + \alpha_2 x^{\gamma}} \quad (x > 1) , \qquad (3.13)$$

as suggested in Ref. 22, with $\gamma = d + 1 = 3$. This fit is represented by the dots in Fig. 8. Trying the value $\gamma = 2d = 4$ for the exponent γ does not result in a good fit. Therefore, our results clearly support the $\gamma = d + 1$ alternative. We refer the reader to Furukawa's²² for a detailed discussion. In three dimensions detailed simulations reviewed in Ref. 1 also show that $\gamma = d + 1$.

We see then that the analytic predictions are clearly verified by the numerical solution of the recursion relations. We have also seen that the logarithmic growth law is not at all in contradiction with the good power-law fits found in MC simulations. Indeed, we believe⁴ that it is likely that many of the experimental or three-dimensional simulation results in which power-law behavior is reported to be found would actually show logarithmic behavior over a sufficiently extended time range.

IV. CONCLUSIONS

We have, in this paper, presented a RG theory of spinodal decomposition in a square-lattice Ising model with Kawasaki dynamics in zero external field. The theory describes self-similar behavior quantitatively. The conservation of the order parameter, the fixed-length sum rule, and the initial time derivative of all the two-point correlation functions are dealt with properly. The time rescaling factor used was obtained in Ref. 4 and is strongly temperature dependent. The main results of the theory are the logarithmic growth law for the typical domain size at long time, which is associated with the freezing of the system for quenches to zero temperature. This is, however, not in conflict with the power-law result of MC simulations. Instead, we have found that in the time region of the MC simulation the logarithmic growth law may be approximated by a power-law fit with an effective exponent. The exponent calculated from our theory is found to be in excellent agreement with that from MC simulations. Our calculations of various other quantities seem to agree well with existing MC simulations.

Here, we can give a physical argument in support of the logarithmic growth law. Let us first recall the nature of the LCA argument,¹¹ at the most heuristic level, for a NCOP. In that case it is argued that at long times the dynamics is dominated by interfacial motion. It is assumed that a typical velocity v of an interface will be driven by the curvature κ . Thus, $v \propto \kappa$, and since $v \approx dL/dt$ and $\kappa \propto L^{-1}$ one immediately obtains $L \propto t^{1/2}$. In the present case the physics is quite different. With exchange dynamics the process of moving particles across an interface becomes increasingly difficult as one lowers the temperature, and we expect the interface velocity to be activated, $v = v_0 \exp(-\kappa Q)$. The kinetics at the interface do depend on the local curvature. In particular, the exchange probability and v are clearly larger for larger local curvature. Thus we expect $Q \propto 1/\kappa - 1/\kappa_0$, where the $1/\kappa_0$ term indicates that Q vanishes for sufficiently large curvature. This takes into account that monomers, which have the largest possible curvature, do propagate at zero temperature, although they are quickly absorbed into larger structures. If we consider again $v \approx dL/dt$, $Q = L - L_0$, and replace K_F by $1/\xi$, we have

$$\frac{dL}{dt} = v_0 e^{-(L-L_0)/\xi} \,. \tag{4.1}$$

The solution to this equation, up to a multiplicative constant, is

$$L = \xi \ln \left[\frac{t v_0}{\xi} \exp(L_0 / \xi) \right], \qquad (4.2)$$

which agrees with our result (3.5) if we take the index $\beta = -1$. The RG result, using $\Delta_0 = 0.41$ at b = 2, would be $\beta = -1.3$. Given the statistical uncertainty in Δ_0 and the heuristic nature of the above argument, the agreement must be considered good. Thus, we have strong evidence that the growth kinetics in this problem involves both the curvature and an activation process at the interface.

Our work here, and in Ref. 4, raises several questions. The first concerns whether this logarithmic growth law we find for a square lattice will extend to three dimensions and other lattices. The argument above would indicate that these results would still apply, but this should be investigated further. We come then to a broader question: In determining the various types of growth in the kinetics of first-order phase transitions, what is the role of the zero-temperature kinetics? There is mounting evidence²³ that zero-temperature freezing, associated with activated kinetics, will influence growth laws at finite temperatures. How do we quantify this notion and use it to classify various systems? Our work suggests that the low-temperature behavior of the rescaling factor Δ is the appropriate quantity to investigate in this respect, but it would be very useful to understand at a more physical level which systems will and which will not freeze. These questions are associated with both the existence of conservation laws and the nature of the ground state, but not in an obvious fashion.

From our renormalization-group point of view the "universal" features of kinetics of first-order phase transitions are governed by the low temperature fixed point. In the case of the LCA growth law this fixed point is associated with the equilibrium ground state. In those circumstances where freezing is possible the system will not equilibrate and the fixed point is not simply associated with the equilibrium ground state.²⁴ In a RG approach we would like to associate this change in the structure of the fixed point with the change of some variable²⁵ from stable to marginal in a RG sense. Variables which may play a role of this type are the concentration (away from 50%) and the density of vacancies (away from zero).

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*Permanent address.

- [†]Present address: Department of Physics and Astronomy, University of Maryland, College Park., MD 20742.
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- ¹⁷See Eqs. (2.4) and (2.7) of Ref. 4. In the discussion below we treat the continuum limit and work in d dimensions. We expect our results will extend to the lattice system in the limit of large distances.
- ¹⁸The factor of v^2 is discussed in Refs. 4 and 9 and arises because the long-time-ordered value of $R_M(t)$ is m_E^2 .
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- ²⁴In the LCA case Δ is a constant and the zero-temperature ground state is allowed to develop freely. In the SE case Δ depends on K_F and leads to a competition with the growing ground state.
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