Instability, spinodal decomposition, and nucleation in a system with continuous symmetry

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The growth kinetics of the time-dependent Ginzburg-Landau model is studied in the large-N limit. Quenches from an initial equilibrium state at infinite temperature to a temperature corresponding to an unstable state beneath the coexistence curve are studied for both a conserved order parameter (COP) and for a nonconserved order parameter (NCOP). In both cases the system grows the order corresponding to the final equilibrium state by growing domains of the new phases. These domains are reflected through the existence of a peak in the structure factors which is evolving toward a Bragg peak as time increases. We find that these peaks satisfy a scaling relation similar to that for the related cases with a scalar order parameter. The characteristic lengths in these scaling laws satisfy a power-law behavior, $L(t)\sim t^a$, where $a=\frac{1}{2}$ for a NCOP and $a=\frac{1}{4}$ for a COP. Riding on top of the developing long-range order are the Nambu-Goldstone modes which, we show, build smoothly into the q^{-2} behavior in the transverse correlation functions. The time evolution of the system after an isothermal field reversal is also studied. The increase in the time characterizing the reversal of the magnetization is found to be approximately inversely proportional to the magnitude of the field reversal. No limit of stability is found.

I. INTRODUCTION

Spontaneous symmetry breaking associated with ordering in systems with a continuous symmetry leads to a number of interesting effects including the existence of Nambu-Goldstone (NG) modes.[1] The word "spontaneous" used alone is, in a certain sense, very misleading. This ordering and the associated NG modes do not just appear as a function of time. They must be *grown*. We discuss here[2] this growth process in the case of the time-dependent Ginzburg-Landau (TDGL) model[3] in the large-N limit,[4] where N is the number of components of the order parameter. In this limit, the problem of the growth of order after a sudden temperature quench or a rapid magnetic field flip can be reduced to an integro-differential equation for a single variable which can be solved numerically to arbitrary accuracy. Thus we can essentially solve the problem exactly. This allows us to investigate the growth kinetics for this model in detail and, as we shall see, follow the build up of NG modes as a function of time after a quench.

There have been significant advances[5] in recent years in our understanding of the growth kinetics of systems suddenly constrained to be in an unstable state. The most important advances have included the identification[6] of scaling behavior in a wide variety of systems and the determination of the growth law as a function of time for the typical domain size $L(t)$, which is the dominant length that produces the scaling behavior. It is now well understood that there are various classes of systems which share similar behaviors. These classes are distinguished first by whether one has a conserved order parameter (COP) or a nonconserved order parameter (NCOP). Then, for example, it is believed that systems with a NCOP and two degenerate ground states (spin-$\frac{1}{2}$ Ising model or a binary alloy, for example)will have a curvature driven growth law, as proposed by Lifshitz,[7] and Cahn and Allen[8] (LCA), of $L(t)\sim t^n$, $n=\frac{1}{2}$. This has been extensively confirmed both experimentally[9] and theoretically.[10] There are also certain "scaling functions" which are shared among members of this set of systems. See Refs. 11 and 12 for a discussion of the degree to which one has "universality" in this case.

For systems with a NCOP but with a ground state of a higher degeneracy, things are not quite as clear. In the case of the q-state clock model,[13] it appears that the growth law is still given by the LCA result. However, in the case of the q-state Potts model it appears[14] that n is a function of q and, indicating some possible problems in a power-law analysis, apparently a function of temperature for low temperatures.

The case of a COP is less well understood. In cases where it is sensible to assume that growth proceeds through the formation of droplets, mobilities are such that one has local equilibrium at an interface, and hydrodynamic interactions are not important, the theory[15] of Lifshitz and Slyozov is applicable, and one obtains a $L\sim t^{1/3}$ growth-law and scaling behavior. There are, however, interesting situations where the assumptions given above are not valid. Mazenko, Valls, and Zhang[16] have recently shown that the growth kinetics of the spin-exchange kinetic Ising model, in the absence of an external field, are dominated by the activated nature of diffusion across an interface at low temperatures. This leads then to an $L\sim \ln t$ growth law at long times. It is also important to note in this case that the morphology[17] of the

growth was not dropletlike but percolative.

If one includes hydrodynamic interactions, as would be present in a fluid, these ultimately should control the growth kinetics (in the absence of gravity) giving[18] $L \sim t$. Clearly, there is a great deal more to understand in the case of a COP since only the case of two degenerate ground states has been extensively studied.

Notice that all of the work mentioned above is for a scalar order parameter; $N=1$ in the language of critical phenomena. The growth kinetics of systems with continuous symmetry ($N>1$) has been virtually unexplored. This is due partly to the historical fact that most work in this area has been on systems of metallurgical interest or on fluids. In both cases the order parameters are typically scalars. There are a variety of situations in condensed-matter physics where the order parameter corresponds to $N>1$ and where we might expect the growth kinetics to differ significantly from those of systems with discrete symmetries. The simplest examples are the growth kinetics of He^4 ($N=2$) quenched below the λ transition or an isotropic antiferromagnet ($N=3$) like $RbMnF_3$ quenched to below the Néel temperature. The N-vector model in the large-N limit is the logical place to start an analysis of the growth kinetics of systems with a continuous symmetry. The model gives a realistic description of a system undergoing a second-order phase transition, and one can define a kinetics with the main features, dissipation and equilibration, which have played a prominent role in the study of the growth kinetics in $N=1$ systems. A key new ingredient in the analysis is the role of Nambu-Goldstone modes in the growth process. This model allows us to deal with this phenomenon in a straightforward and believable fashion. One deficiency of this model is that it does not have any "defects," such as vortices, which we know can play a role[19] in X-Y and Heisenberg models.

II. THE MODEL

A. Statics

We consider a system, in D spatial dimensions, described by the Landau-Ginzburg-Wilson free-energy functional

$$F[\boldsymbol{\phi}]=\tfrac{1}{2}\int d^Dx[(\nabla\boldsymbol{\phi})^2+r\boldsymbol{\phi}^2+u(\boldsymbol{\phi}^2)^2/2N-2\mathbf{H}\cdot\boldsymbol{\phi}]\ , \tag{2.1}$$

where $\boldsymbol{\phi}(\mathbf{x})$ is an N-component order parameter

$$\boldsymbol{\phi}(\mathbf{x})=(\boldsymbol{\phi}_1(\mathbf{x}),\ldots,\boldsymbol{\phi}_N(\mathbf{x}))\ . \tag{2.2}$$

As is customary in the theory of critical phenomena, $F[\boldsymbol{\phi}]$ must be regarded as a reduced free energy, with the temperature absorbed into the coupling constants. Furthermore, we keep u fixed and positive and we study the behavior of the system under variations of r and $\mathbf{H}$. In equilibrium, averages correspond to functional integrals over $\boldsymbol{\phi}$ weighted by e^{-F}. Taking the external field directed along the 1 axis,

$$H_i=H\delta_{i,1}\ ; \tag{2.3}$$

the average order parameter is of the form

$$\langle\phi_i(\mathbf{x})\rangle=M\delta_{i,1}\ . \tag{2.4}$$

Introducing the new fluctuating variable $\delta\phi_i=\phi_i-\langle\phi_i\rangle$, the longitudinal and transverse correlation functions are defined by

$$C_{||}(\mathbf{x}-\mathbf{x}')=\langle\delta\phi_1(\mathbf{x})\delta\phi_1(\mathbf{x}')\rangle \tag{2.5}$$

and

$$C_{\perp}(\mathbf{x}-\mathbf{x}')=\langle\delta\phi_i(\mathbf{x})\delta\phi_i(\mathbf{x}')\rangle,\quad i\neq 1\ . \tag{2.6}$$

In the large-N limit,[20] the equilibrium behavior of the system is completely described by $M, C_{||}$ and $C_{\perp}$. These quantities obey the set of equations

$$[r+u(S+m^2)]m=h\ , \tag{2.7}$$

$$C_{\perp}(q)=\frac{1}{q^2+K_{\perp}^2}\ , \tag{2.8}$$

$$C_{||}(q)=\frac{1}{q^2+K_{||}^2}\ , \tag{2.9}$$

where $m=M/\sqrt{N}$ and $h=H/\sqrt{N}$. The static structure factors $C_{\perp}(q)$ and $C_{||}(q)$ are the Fourier transforms of $C_{\perp}(\mathbf{x}-\mathbf{x}')$ and $C_{||}(\mathbf{x}-\mathbf{x}')$, and the inverse correlation lengths $K_{\perp}, K_{||}$ are given by

$$K_{\perp}^2=r+u[S+m^2]\ , \tag{2.10}$$

$$K_{||}=r+u[S+3m^2]\ , \tag{2.11}$$

with

$$S=\int\frac{d^Dq}{(2\pi)^D}C_{\perp}(q)\ . \tag{2.12}$$

From the equation of state (2.7) we have for $h=0$ the solutions

$$m=0,\quad r>r_c\ , \tag{2.13a}$$

$$m^2=\frac{(r_c-r)}{u},\quad r<r_c \tag{2.13b}$$

where $r_c=-uS_c$ is the critical point value of r and

$$S_c=\int\frac{d^Dq}{(2\pi)^D}\frac{1}{q^2}\ . \tag{2.14}$$

Above the critical point there is no distinction between longitudinal and transverse modes:

$$C_{||}(q)=C_{\perp}(q)=\frac{1}{q^2+K^2}\ , \tag{2.15}$$

with $K^2=r+uS$. On the other hand, below the critical point, from Eqs. (2.13b), (2.7), (2.10), and (2.11), there is spontaneous symmetry breaking and the transverse correlation length vanishes for all $r<r_c$, indicating the existence of Nambu-Goldstone modes in the transverse directions. Correspondingly the structure factors for $r<r_c$ are given by

$$C_{||}(q)=\frac{1}{q^2+2(r_c-r)}\ , \tag{2.16}$$

$$C_\perp(q)=\frac{1}{q^2}\ , \tag{2.17}$$

and from the latter equation we have

$$S\equiv S_c,\ \text{ for all } r<r_c\ . \tag{2.18}$$

Finally, we recall the results of the free theory, obtained from the previous results by setting $u=0$. In the absence of an external field, the structure factor is given by

$$C_0(q)=\frac{1}{q^2+r}\ . \tag{2.19}$$

The critical point is at $r_{0c}=0$, and the equilibrium state is not defined for $r<r_{0c}$.

B. Dynamics

The dynamics of the model is governed by the time-dependent Ginzburg-Landau model

$$\frac{\partial\boldsymbol{\phi}(\mathbf{x},t)}{\partial t}=-\Gamma\frac{\delta F}{\delta\boldsymbol{\phi}}+\boldsymbol{\eta}(\mathbf{x},t)\ , \tag{2.20}$$

where Γ is the kinetic coefficient and $\boldsymbol{\eta}(\mathbf{x},t)$ is a Gaussian white noise satisfying

$$\langle\boldsymbol{\eta}(\mathbf{x},t)\rangle=0\ , \tag{2.21a}$$

$$\langle\eta_i(\mathbf{x},t)\eta_j(\mathbf{x}',t')\rangle=2\Gamma(\mathbf{x})\delta_{ij}\delta(\mathbf{x}-\mathbf{x}')\delta(t-t')\ . \tag{2.21b}$$

We study the dynamics with conservation of the order parameter,

$$\Gamma(\mathbf{x})=-\Gamma\nabla^2\ , \tag{2.22}$$

and without conservation of the order parameter,

$$\Gamma(\mathbf{x})=\Gamma\ . \tag{2.23}$$

Introducing the time-dependent magnetization

$$\langle\phi_i(\mathbf{x},t)\rangle=M(t)\delta_{i,1} \tag{2.24}$$

and the fluctuating fields

$$\delta\phi_i(\mathbf{x},t)\equiv\phi_i(\mathbf{x},t)-\langle\phi_i(\mathbf{x},t)\rangle\ , \tag{2.25}$$

the equation of motion (2.20), in the large-N limit,[21] yields the set of equations

$$\frac{\partial m}{\partial t}=\Gamma(\mathbf{x})(\zeta_i m+h)\ , \tag{2.26}$$

$$\frac{\partial(\delta\phi_i)}{\partial t}=\Gamma(\mathbf{x})(\nabla^2+\zeta_i)\delta\phi_i+\eta_i(\mathbf{x},t)\ , \tag{2.27}$$

where

$$\zeta_i=-(r+uS+m^2+2m^2\delta_{i,1}) \tag{2.28}$$

and

$$S=\langle\delta\phi_i^2(\mathbf{x},t)\rangle,\ \ i\neq1\ . \tag{2.29}$$

The quantities of interest, besides the magnetization $m(t)$, are the quasistatic correlation functions

$$C_i(\mathbf{x}-\mathbf{x}',t)=\langle\delta\phi_i(\mathbf{x},t)\delta\phi_i(\mathbf{x}',t)\rangle\ . \tag{2.30}$$

Using (2.27) and Fourier transforming, we obtain the equation of motion for the corresponding structure factors

$$\frac{\partial C_i(\mathbf{q},t)}{\partial t}=-2\Gamma(\mathbf{q})(q^2-\zeta_i)C_i(\mathbf{q},t)+2\Gamma(\mathbf{q})\ , \tag{2.31}$$

where

$$\Gamma(\mathbf{q})=\Gamma\ \ (\text{NCOP})\ , \tag{2.32a}$$

$$\Gamma(q)=\Gamma q^2\ \ (\text{COP})\ , \tag{2.32b}$$

and $S(t)$, defined by Eq. (2.29), is given by

$$S(t)=\int\frac{d^Dq}{(2\pi)^D}C_\perp(\mathbf{q},t)\ . \tag{2.33}$$

The set of equations (2.26), (2.28), (2.31), and (2.33) define the dynamics. In order to have dimensionless equations, let us measure lengths in units of Λ^{-1}, where Λ is a momentum cutoff, and time is in units of $2\Gamma\Lambda^2$ for a NCOP and $2\Gamma\Lambda^4$ for a COP. We then have the set of equations

$$\frac{\partial m}{\partial t}=\tfrac{1}{2}(\zeta_\perp m+h)\ \ (\text{NCOP})\ , \tag{2.34}$$

$$\frac{\partial m}{\partial t}=0\ \ (\text{COP})\ ,$$

$$\frac{\partial C_i(\mathbf{q},t)}{\partial t}=-(q^2-\zeta_i)C_i(\mathbf{q},t)+1\ \ (\text{NCOP})\ , \tag{2.35}$$

$$\frac{\partial C_i(\mathbf{q},t)}{\partial t}=-q^2(q^2-\zeta_i)C_i(\mathbf{q},t)+q^2\ \ (\text{COP})\ , \tag{2.36}$$

with

$$\zeta_i=-(r+S+m^2+2m^2\delta_{i,1})\ , \tag{2.37}$$

which is obtained after choosing the coupling constant u such that $u\Lambda^{D-4}=1$ and

$$S=K_D\int_0^1 dq\,q^{D-1}C_\perp(\mathbf{q},t)\ , \tag{2.38}$$

$$K_D=2^{1-D}\pi^{-D/2}/\Gamma(D/2)\ . \tag{2.39}$$

In the following we shall be interested in the nonequilibrium processes arising when the system, initially prepared in an equilibrium state, undergoes an instability through a sudden change of the thermodynamic parameters. The most general process of this kind is given by $(r_I,h_I)\rightarrow(r_F,h_F)$, where (r_I,h_I) characterize the initial equilibrium state ($t\leq t_0$), and (r_F,h_F) are the new values (at $t=t_0$) characterizing the bath that drives the system to final equilibrium state at "temperature" r_F and in a field h_F. In this paper the following particular cases will be analyzed: (i) quench in zero field for the NCOP

$$(r_I,h_I=0)\rightarrow(r_F<r_I,h_F=0)\ ,$$

(ii) quench for a COP with a constant magnetization

$$(r_I,h_I\neq0)\rightarrow(r_F<r_I,h_F=0)\ ,$$

and (iii) isothermal field reversal

$$(r_I<r_c,h_I=0^+)\rightarrow(r_F=r_I,h_F<0)\ .$$

The numerical calculations will be carried out for $D=3$, where $r_c=-S_c=-1/2\pi^2=-0.051$.

III. NCOP: TEMPERATURE QUENCH IN ZERO FIELD

In this section we consider the time development of the instability when the system, initially prepared in the state $(r_I, h_I=0)$, evolves toward the new equilibrium state characterized by $(r_F<r_I, h_F=0)$. We choose $r_I=+\infty$ and we will consider both $r_F>r_c$ and $r_F<r_c$. The very large value of r_I produces a very narrow one-well potential which constrains the order parameter to remain near the origin with no fluctuations (see Fig. 1). Consequently, we have

$$C(\mathbf{q},t_0)=0\ , \tag{3.1a}$$

$$S(t_0)=0\ . \tag{3.1b}$$

A. Noninteracting theory

Before proceeding with the full analysis of the dynamics associated with Eqs. (2.34)—(2.37), it is worth discussing the noninteracting theory ($u=0$), since it gives some insight into the importance of nonlinearities in the problem. In this limit each mode evolves independently from the others through the equation

$$\frac{\partial C_0(\mathbf{q},t)}{\partial t}=-(q^2+r_F)C_0(\mathbf{q},t)+1\ , \tag{3.2}$$

which has the solution (for $r_I>0$)

$$C_0(\mathbf{q},t)=C_0(\mathbf{q},r_F)+e^{-(q^2+r_F)t}(C_0(\mathbf{q},r_I)-C_0(\mathbf{q},r_F))\ , \tag{3.3}$$

where $C_0(\mathbf{q},r)$ is given by (2.19). For $r_F>0$, one has exponential relaxation to the equilibrium state $C_0(\mathbf{q},r_F)$. For $r_F<0$, however, all modes with $q<q_c=|r_F|^{1/2}$ are intrinsically unstable and the system will not equilibrate. The distinction between the two cases is illustrated in Fig. 2 through the time development of

$$S_0(t)=\int\frac{d^3q}{(2\pi)^3}C_0(\mathbf{q},t)\ .$$

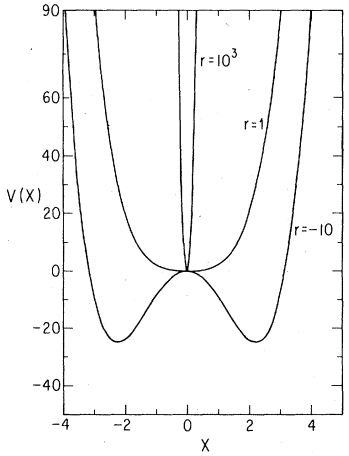

FIG. 1. Potential $V(x)=rx^2+x^4$ for various values of r.

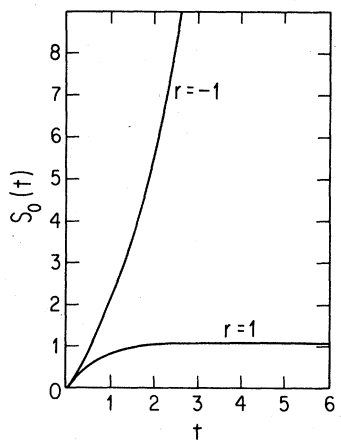

FIG. 2. Time evolution of $S_0(t)$ in the noninteracting theory for quenches above and below $r_{0c}=0$.

B. Interacting theory: $r_F\geq r_c$

The interacting theory can be solved numerically using a standard forward step time integration and a numerical integration over a 100-point mesh in q to obtain $S(t)$. Above the critical point ($r_F>r_c$), the time evolution in the interacting case is not expected to be qualitatively different from the noninteracting case. The behavior of $S(t)$ (Fig. 3) displays the same saturation pattern observed in the noninteracting case for $r>0$. In this case, since $r_c=-1/2\pi^2<0$, there is also no instability for $r_c<r<0$, as shown in Fig. 3 for $r_F=-0.025$.

The structure factor $C(\mathbf{q},t)$ shown in Fig. 4 displays a uniform growth from the initial value (3.1) to the final equilibrium Lorentzian form (2.15). The same behavior is observed for a quench to the critical point $r_F=r_c$, which for completeness is shown in Fig. 5. One finds that at the peak height a $q=0$ grows linearly with time and the width narrows with a $t^{-1/2}$ behavior.

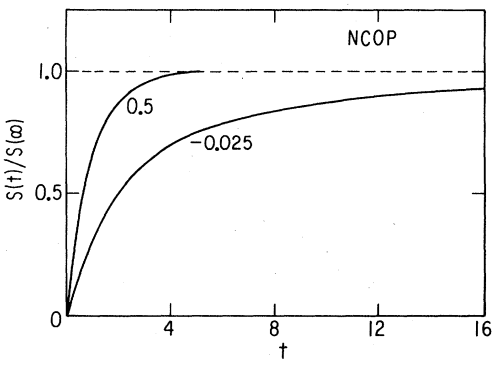

FIG. 3. Time evolution of $S(t)$ for NCOP in the interacting theory for a quench above the critical point ($r_F=0.5$ and $r_F=-0.025$).

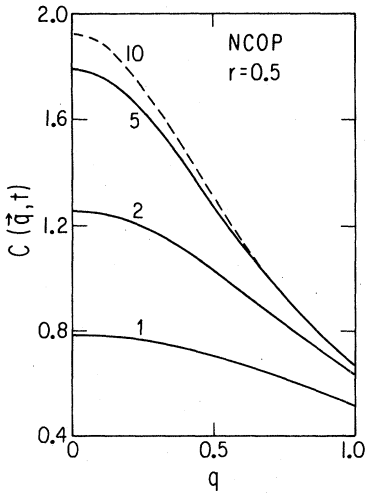

FIG. 4. Time evolution of the structure factor for NCOP above the critical point ($r_F=0.5$). The equilibrium structure factor given by Eq. (2.15) (dashed curve) is reached for $t\sim 10$.

C. Interacting theory: $r_F < r_c$

As was discussed in Sec. II, in the equilibrium state with $r_F < r_c$ the symmetry is broken and the system develops a spontaneous magnetization, (2.13b) with $u=1$,

$$m^2 = r_c - r_F \ , \tag{3.4}$$

and the static structure factor behaves differently in the longitudinal and transverse directions as indicated in Eqs. (2.16)—(2.17). Therefore, the time evolution after a quench with $r_F < r_c$ is expected to exhibit a rich and complicated behavior. The system starts out in an initial symmetric equilibrium state and has to comply with the requirements of a final equilibrium state, which requires symmetry breaking, through dynamical equations which do not contain a symmetry-breaking mechanism. Therefore, for all finite times there is no spontaneous magnetization $[m(t)=0]$ and there is no distinction between longitudinal and transverse modes. The equation of motion is given by

$$\frac{\partial C_i(\mathbf{q},t)}{\partial t} = -(q^2-\zeta)C_i(\mathbf{q},t)+1 \ , \tag{3.5}$$

with

$$\zeta = -(r_F+S) \ . \tag{3.6}$$

For very early times we expect, given the initial condition (3.1), that $\zeta \sim -r_F$ and the dynamics will proceed as in the noninteracting case. Namely, that each mode will evolve independently from the others, the modes with $q<q_c=|r_F|^{1/2}$ being unstable. We find, in particular for $r_F=-10$, that the growth of the peak at $\mathbf{q}=0$ is accurately given by (3.3) for times up to $t=0.9$. For larger times, however, the noninteracting theory rapidly breaks down and nonlinear interactions become increasingly important. It is precisely these nonlinear interactions which tame the instability and eventually equilibrate the system. This is due to the feedback of the fluctuations on the amplification factor, given by the presence of S in Eq. (3.6). As the instability proceeds, the fluctuations grow and reduce the value of ζ, i.e., of the characteristic wave vector $q_c=\zeta^{1/2}$. Since the expected equilibrium state is given by Eqs. (2.16), (2.17), and (2.18), and it must correspond to the vanishing of the time derivative in the left-hand side of Eq. (3.5), we expect that equilibrium is reached as ζ vanishes and $S(t)$ grows toward the saturation value $S(\infty)=|r_F|$. This is clearly illustrated in Fig. 6, which gives the behavior of $S(t)/S(\infty)$ for different values of r_F well below the critical point. One can clearly recognize two different time regimes: the fast transient previously discussed at the early stage of the instability, which brings $S(t)$ very close to the saturation value $S(\infty)=|r_F|$, and the ensuing slow transient for subsequent times. It is with this latter time evolution that we will be mainly concerned in this paper. The mechanism governing the instability in the slow transient is quite different from the one operat-

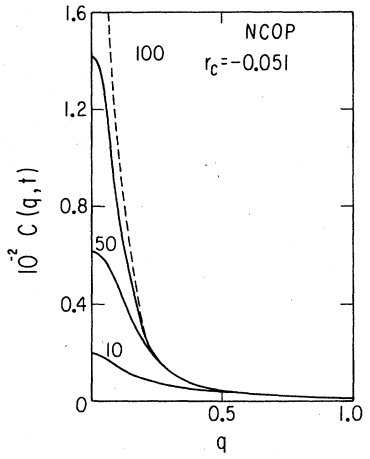

FIG. 5. Time evolution of the structure factor for NCOP and a quench at the critical point ($r_F=r_c=-0.051$). The dashed curve represents the asymptotic critical structure factor $C(\mathbf{q},\infty)=1/q^2$.

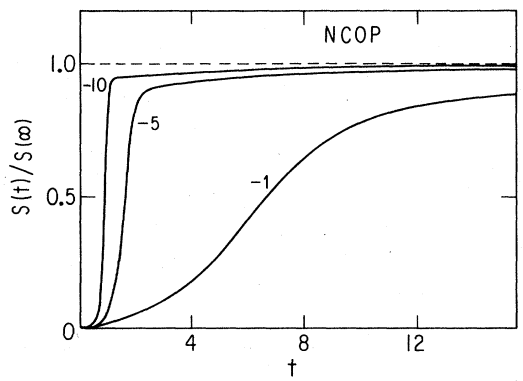

FIG. 6. Time evolution of $S(t)$ for NCOP and various quenches below the critical point ($r_F=-1$, $r_F=-5$, $r_F=-10$).

ing at the early stage and, in fact, we expect it to be dominated by the interaction among the modes.

Notice that although ζ is small and $S(t)\sim S(\infty)$, this is still a situation far from equilibrium, since Eq. (2.18) requires in the equilibrium state $S=S_c=-r_c \ll S(\infty) = |r_F|$. In other words, during the early stage the system has managed to reduce the amplification factor by developing anomalously large fluctuations. The dynamics of the later stages must then satisfy the twofold requirement of keeping the amplification factor small (ζ vanishes as t^{-1}) and reducing the value of $S(\infty)$ to S_c. This is done by shifting the fluctuations from the high-q modes to the low-q modes through the mode interaction, as is illustrated in Fig. 7, which depicts the time evolution of the structure factor. In the early stage, $C(\mathbf{q},t)$ grows from zero almost uniformly for all values of q shown (Fig. 7 corresponds to $|r_F|=10$ and therefore $q_c>1$, initially). Subsequently, after $S(t)$ has reached a value close to $S(\infty)$, the dynamics of the structure factor evolves via the relaxation of the higher-q modes to the asymptotic equilibrium value $\sim 1/q^2$ (dashed curve in Fig. 7) and by the continuing growth of the low-q modes. Eventually, when ζ is very close to 0 and $S(t)$ to $S(\infty)$, only the $q=0$ mode keeps on growing without limit, while $C(\mathbf{q},t)$ approaches the Nambu-Goldstone contribution $C_\perp(\mathbf{q})=1/q^2$ for $q\neq 0$.

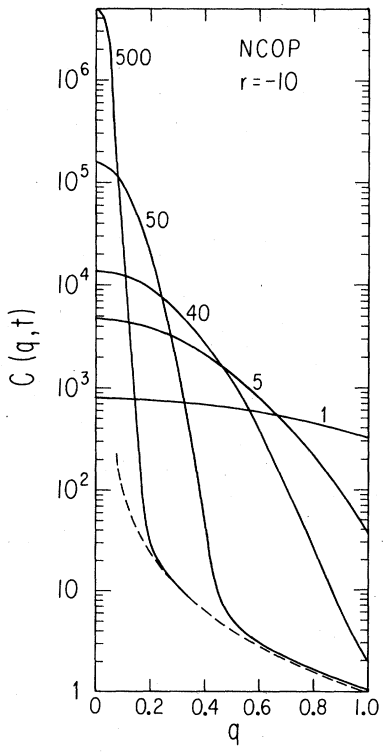

FIG. 7. Time evolution of the structure factor for NCOP and a quench below the critical point ($r_F=-10$). The dashed curve represents the equilibrium Nambu-Goldstone contribution $C_{\rm NG}(\mathbf{q})=1/q^2$.

This kind of behavior means that the system attempts to reduce the anomalous high value of fluctuations [as represented by $S(t)\sim S(\infty)$] by ordering. The ordering corresponds to the growth of domains of the desired final equilibrium state. However, since there is no dynamical mechanism for symmetry breaking in the equations of motion, all possible orientations of the ordering will be represented among the domains, and $m(t)$ remains zero on averaging over the entire system. The development and inexorable growth of these domains is reflected in the form of the structure factor. The solution of the equation of motion (3.5), with the initial conditions (3.1) is given by

$$C(\mathbf{q},t)=\int_{t_0}^{t} dt' \exp\left[-\int_{t'}^{t} ds(q^2-\zeta)\right], \tag{3.7}$$

which can also be rewritten as

$$C(\mathbf{q},t)=C(\mathbf{q},\bar{t})\exp\left[-\int_{\bar{t}}^{t} ds(q^2-\zeta)\right] + \int_{\bar{t}}^{t} dt' \exp\left[-\int_{t'}^{t} ds(q^2-\zeta)\right], \tag{3.8}$$

where $\bar{t}$ is an instant-of-time intermediate between t_0 and t. If $\bar{t}$ is chosen in the late stage of the relaxation then, as it has been remarked above, ζ is small and there is a range of wave vectors $q^2 \gg \zeta$, such that ζ can be neglected in the right-hand side of Eq. (3.8), yielding

$$C(\mathbf{q},t)=C(\mathbf{q},\bar{t})e^{-q^2(t-\bar{t})}+[1-e^{-q^2(t-\bar{t})}]/q^2 . \tag{3.9}$$

Furthermore, if $(t-\bar{t})$ is sufficiently large, the first term can be neglected with respect to the second one. This holds true as long as $q^2>\zeta$. On the other hand, for $q^2 \lesssim \zeta$ (which gives a narrow interval around the origin when $\bar{t}$ is chosen very large) the opposite occurs, namely, both terms in the right-hand side of Eq. (3.8) describe exponential growth but the first one is overwhelmingly bigger since it contains the cumulative effect of the instability at the earlier times. Hence, in the late stage the structure factor to a good approximation can be written in the form

$$C(\mathbf{q},t)=D(\mathbf{q},t)+C_{\rm NG}(\mathbf{q},t) , \tag{3.10}$$

where

$$C_{\rm NG}(\mathbf{q},t)=[1-e^{-q^2t}]/q^2 , \tag{3.11}$$

$$D(\mathbf{q},t)=D(\mathbf{q},\bar{t})\exp\left[-\int_{\bar{t}}^{t} ds[q^2-\zeta]\right] , \tag{3.12}$$

and

$$C(\mathbf{q},t)\sim \begin{cases} D(\mathbf{q},t), & q^2 \lesssim \zeta \\ C_{\rm NG}(\mathbf{q},t), & q^2>\zeta . \end{cases} \tag{3.13}$$

The contribution (3.11) describes the growth of the Nambu-Goldstone modes and yields asymptotically the $1/q^2$ behavior. The term $D(\mathbf{q},t)$ instead describes the growth of the central Bragg peak associated with the development of order. Furthermore, this latter contribution can be put in scaling form,

$$D(\mathbf{q},t)=AL^3(t)F(\mathbf{q}L(t)) , \tag{3.14}$$

with $F(0)=1$, implying that at this stage of the evolution all the relevant time dependence appears through one fun-

damental length $L(t)$, which gives the linear dimension of the domains. The form (3.14) of $D(\mathbf{q},t)$ is immediately obtained from Eq. (3.12) after defining $F(x)$ through

$$D(\mathbf{q},t)=D(\mathbf{q}=0,t)F(\mathbf{q},t) .$$

This gives

$$F(\mathbf{q},t)=F(\mathbf{q},\bar{t})e^{-q^2(t-\bar{t})}$$

with the solution $F(\mathbf{q},t)=e^{-q^2t}$, which can be rewritten as

$$F(x)=e^{-ax^2} , \tag{3.15}$$

with $x=qL(t)$ and

$$L(t)=(t/a)^{1/2} . \tag{3.16}$$

Defining $L^{-1}(t)$ as the width at half-height of the peak, we have $a=\ln 2$ and $L(t)=1.20t^{1/2}$. Even though $m(t)=0$ at all times, the quantity A in Eq. (3.14) evolves to the value $A=m_F^2$ given by Eq. (3.4), where m_F is the value of the spontaneous magnetization associated with the final equilibrium state.

The above predictions for the structure factor are substantiated by the numerical computation. In fact, Eqs. (3.10), (3.11), and (3.14) give an accurate representation of the numerical data for the structure factor in the late stage. After the subtraction of the Nambu-Goldstone contribution (3.11), the Gaussian (3.15) with $a=\ln 2$ gives an excellent fit of the data for the shape function $x<4$, and it is found to be independent of time and of the value of r_F (Fig. 8). From the data for the inverse of the width of the Bragg peak, we find $L(t)=1.32t^{0.47}$ for long times, in agreement with Eq. (3.16). We remark that the behavior of $L(t)$ is in agreement with the Lifshitz–Cahn-Allen curvature-driven growth law developed for scalar order parameters. This suggests that in the NCOP case the growth law is independent of the number N of the order-parameter components. The $\mathbf{q}=0$ component of the structure factor satisfies, for long times,

$$C(\mathbf{0},t)=t+423.7t^{3/2} , \tag{3.17}$$

where the $t^{3/2}$ term corresponds to the setting up of domains of the new phases, and the linear term corresponds to the development of Nambu-Goldstone modes within these domains. Finally, notice that the form (3.10) of the structure factor applies to all i components of the order parameter. Namely, the Bragg peak develops along all directions as domains pointing in all directions are formed, and globally the symmetry is not broken.[22] Therefore, the Nambu-Goldstone modes must be present for all values of i, since a given value of i is transverse with respect to all the remaining ones.

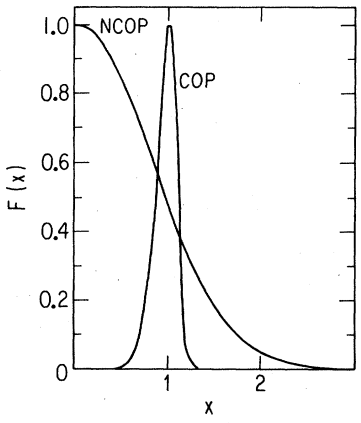

FIG. 8. Shape functions for NCOP and COP.

IV. COP: TEMPERATURE QUENCH WITH CONSTANT MAGNETIZATION

A. Noninteracting case

The noninteracting solution ($u=0$) for a COP corresponds to the Cahn-Hilliard[23]–Cook[24] theory with a structure factor given by (3.3) but with $(q^2+r_F)t$ multiplied by q^2 in the exponential. For $r_F>0$, we obtain exponential relaxation to equilibrium at r_F, except for the point $\mathbf{q}=0$, which is pinned at the initial value $C_0(0,r_I)=1/r_I$ by the conservation law. For $r_F<0$, we again have unstable, exponentially growing modes for wave numbers $q<(|r_F|)^{1/2}$. Because of the conservation law, there is a single wave number $q_M=(|r_F|/2)^{1/2}$ which grows faster than the others. Therefore, $C(\mathbf{q},t)$ is characterized by a peak located at $q=q_M$, which grows exponentially as

$$C(\mathbf{q}_M,t)=2(e^{|r_F|^2t/4}-1)/|r_F| .$$

B. Interacting case

The preparation of the system in the interacting case is a bit more subtle. We assume that in the initial state $(r_I,h_I\neq 0)$ there is a finite magnetization $m_I\neq 0$. Then we imagine suddenly dropping r to a value r_F below the coexistence curve and adjusting the field so that the average magnetization is fixed at the value m_I. On and below the coexistence curve the field vanishes, so that the final equilibrium state is characterized by $(r_F,h_F=0)$. Since in this problem the symmetry is broken from the onset, we must distinguish between longitudinal and transverse modes. We must solve Eq. (2.36) for the transverse direction

$$\frac{\partial C_\perp(\mathbf{q},t)}{\partial t}=-q^2(q^2-\zeta_\perp)C_\perp(\mathbf{q},t)+q^2 , \tag{4.1}$$

where

$$\zeta_\perp=-(r_F+S+m^2) . \tag{4.2}$$

The behavior of $C_{||}(\mathbf{q},t)$ then is obtained from the equation analogous to (4.1) replacing $\zeta_\perp$ with $\zeta_{||}=\zeta_\perp-2m^2$.

In the initial state with $r_I=+\infty$, we have, as in Eqs. (3.1) and (3.2), $C_I(\mathbf{q})=S_I=0$. The first thing we note in this case is that we can map the problem for the transverse modes with a finite magnetization into one with zero magnetization if we replace $r_F+m^2=r_F'$ and treat r_F' as an effective "temperature." Clearly the main qualitative effect of m^2 is to define the coexistence curve.

Therefore, the system is unstable for $r_F' < r_c$. Numerical solution of (4.1), (4.2), and (2.33) proceeds just as in the NCOP case. The behavior of $S(t)$ (Fig. 9) exhibits two time regimes as in the NCOP case, although the transition between the two is smoother and the initial transient is slower, as a consequence of the conservation constraint.

Looking at the structure factor there is an early time region where the noninteracting theory applies, but this is very narrow. As shown in Fig. 10, the structure is characterized by a peak at a finite wave number q_M, but this wave number is time dependent (unlike in the noninteracting theory) and moving toward small values with increased time. The noninteracting prediction that the peak height increases as $e^{|r_F|^2 t/4}$ must break down at very early times since for $t=20$, $C(q_M,t)\sim 10^3$ for $|r_F|=0$. The correct physical picture is that the system again attempts to build a Bragg peak at the zero wave number reflecting the developing order. The conservation law, however, pins the $\mathbf{q}=0$ component at its value, and the best the system can do is build a peak at the finite wave number which moves monotonically toward the origin. This is just the usual process of spinodal decomposition. The new aspect of our problem here is that the system is also attempting to build up Nambu-Goldstone modes which "ride" on the order.

The structure factor in the late stage can be analyzed in the same way as in the NCOP case. We write the solution of Eq. (4.1) in the form analogous to (3.8),

$$C_\perp(\mathbf{q},t)=C_\perp(\mathbf{q},\bar{t})\exp\left[-q^2\int_{\bar{t}}^{t} ds(q^2-\zeta_\perp)\right] + q^2\int_{\bar{t}}^{t} dt' \exp\left[-q^2\int_{t'}^{t} ds(q^2-\zeta_\perp)\right] . \quad (4.3)$$

Since we find numerically that for large times, $\zeta_\perp \sim t^{-1/2}$, for $\bar{t}$ sufficiently large we may write, as in Eq. (3.10),

$$C_\perp(\mathbf{q},t)=D(\mathbf{q},t)+C_{\rm NG}(\mathbf{q},t) , \quad (4.4)$$

with

$$C_{\rm NG}(\mathbf{q},t)=(1-e^{-q^4 t})/q^2 \quad (4.5)$$

and

$$D(\mathbf{q},t)=D(\mathbf{q},\bar{t})\exp\left[-q^2\int_{\bar{t}}^{t} ds(q^2-\zeta_\perp)\right] . \quad (4.6)$$

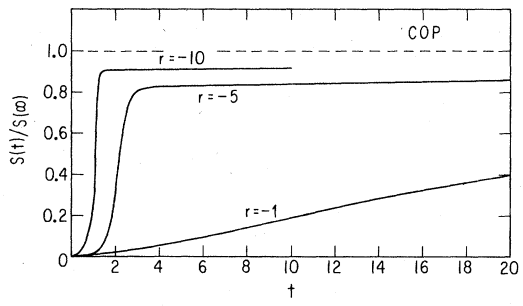

FIG. 9. Time evolution of $S(t)$ for COP and various quenches below the critical point with zero magnetization ($r_F=-1$, $r_F=-5$, $r_F=-10$).

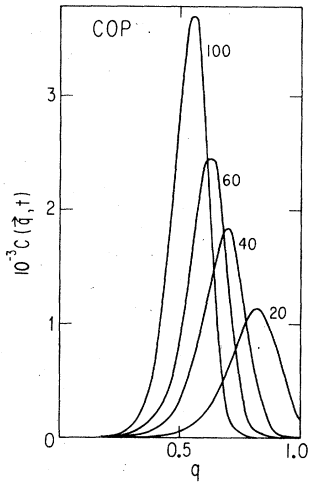

FIG. 10. Time evolution of the structure factor for COP and a quench below the critical point ($r_F=-10$) and zero magnetization.

The peak height and position of the Nambu-Goldstone contribution are given by $P_{\rm NG}=2.01t^{1/2}$ and $q_{\rm NG}=0.6t^{-1/4}$. After subtracting the Nambu-Goldstone contribution, the data for the peak term $D(\mathbf{q},t)$ are well fitted by the scaling form

$$D(\mathbf{q},t)=AL^3(t)F(\mathbf{q}L(t)) , \quad (4.7)$$

with the position of the Bragg peak given by $q_m(t)=L^{-1}(t)=1.7t^{-1/4}$ and the peak height given by $P_M(t)=104.0t^{3/4}$, where the coefficients are for $r_F=-10$. Again we find ($u=1$)

$$A=r_F-m^2-S_c=m_F^2-m^2 , \quad (4.8)$$

and the shape function shown in Fig. 8 is independent of time and of the value of r_F. For $x>2$ the shape function is well approximated by the form[25] $(a+bx^4)^{-1}$, where $a=-1.3\times10^4$ and $b=6.1\times10^3$. The evolution of $C_{||}(\mathbf{q},t)$ in this case is straightforward since $\zeta_{||}=\zeta_\perp-2m^2$, and for large times $|\zeta_\perp|=2m^2$, $\zeta_{||}$ is negative and the driving force $q^2(q^2-\zeta_{||})$ for $C_{||}(\mathbf{q},t)$ is positive and it will relax exponentially to its final equilibrium value.

In summary, we arrive at the following picture: if r_F is below the coexistence curve and $h_F=0$, the equation of state $r_F+S_c+m_F^2=0$ gives, in equilibrium, $m_F^2>m_I^2$. On the other hand, as a consequence of the order-parameter conservation, $m(t)=m_I$. This can be reconciled with the equilibrium requirement by developing order in the perpendicular directions. Then we see that there is a contribution m_I^2 to the total magnetization squared from the longitudinal component and a contribution $m_F^2-m_I^2$ from the "Bragg peaks" in the transverse directions [Eq. (4.8)], yielding the correct equilibrium sum m_F^2.

V. ISOTHERMAL FIELD REVERSAL

In this section we consider a different type of instability, in the case of an NCOP, characterized by the relaxation toward a new equilibrium state through the mecha-

nism of nucleation. We point out that we use nucleation in a generalized sense here because we expect a qualitative difference between the $N=1$ case, where one has sharp interfaces between phases, and $N>1$ where one has "gradual" Bloch walls separating domains. We assume that the system is prepared in an initial equilibrium state lying on the coexistence curve, namely, $r_I<r_c$, with a nonvanishing magnetization

$$m_I=(r_c-r_I)^{1/2} \tag{5.1}$$

pointing in the positive 1 direction. We may think that this state is realized in the presence of a positive and infinitesimally small external field $h_I=0^+$. At some time t_0 the field is suddenly reversed to a negative final value $h_F<0$. The time evolution toward the new equilibrium is governed by Eqs. (2.34) and (2.35) for the magnetization and the transverse structure factor, namely, we must solve (2.34), (2.35), (2.37), and (2.38) with i being a perpendicular component for $m(t)$ and $C_\perp(\mathbf{q},t)$.

The longitudinal structure factor $C_{||}(\mathbf{q},t)$ then is obtained by replacing $\zeta_\perp$ with

$$\zeta_{||}=-(r_I+S+3m^2)\ .$$

The resulting equations have been studied numerically and the behavior of the magnetization, shown in Fig. 11 for $r_I=-10$, exhibits the typical behavior expected when the new phase is formed through a nucleation process. We notice that there is no limit of stability as has been found in certain approximate treatments[26,17] of the $N=1$ case.

Introducing the relaxation function

$$\phi(t)=\frac{m(t)-m_F}{m_I-m_F}\ , \tag{5.2}$$

where m_F is the final equilibrium magnetization satisfying the equation of state (2.7), and defining the lifetime τ of the initial metastable state by

$$\phi(\tau)=\tfrac{1}{2}\ , \tag{5.3}$$

we obtain τ as a function of h_F for $r_F=-10$, as shown in Fig. 12. Our best fit to the data is given by $25.66\,|h_F|^{-1.07}$, while a fit to $\tau\sim|h_F|^{-1}$ fits well for $|h_F|<1$, but less well for higher values of $|h_F|$.

Of particular interest is the behavior of the transverse

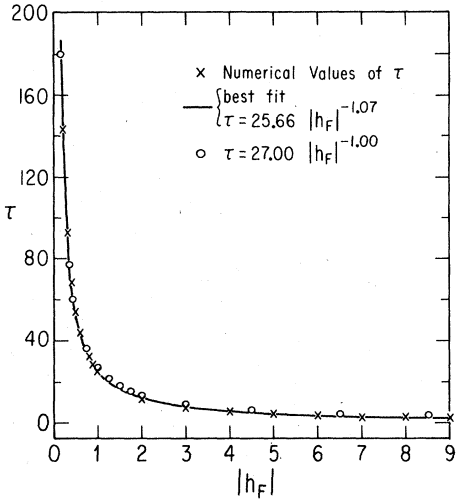

FIG. 12. Dependence of τ [see Eq. (5.3)] on $|h_F|$ for $r_I=-10$. $\times$ are the result of our numerical solution for $m(t)$. The solid curve is the fit to $\tau=25.66\,|h_F|^{-1.07}$ and the open circles are for $\tau=27.00\,|h_F|^{-1}$.

structure factor during the time evolution. Initially $\zeta_\perp=0$, and in the transverse directions there is only the contribution of the Nambu-Goldstone modes $C_\perp(q,t_I)=1/q^2$. The effect of the negative external field h_F in the right-hand side of Eq. (2.34) is to reduce the size of the magnetization, which in turn produces a positive value of $\zeta_\perp$ and therefore a growth of the fluctuations in the transverse directions. These reach a maximum when the magnetization along the longitudinal direction goes through zero and then decrease gradually as the magnetization grows negative (Fig. 13). The time development of the structure factor is illustrated in Fig. 14 for $h_F=-1.0$ and $r=-10$. The continuous lines show the growth of the structure factor above the $1/q^2$ behavior as the magnetization decreases from the initial value to the zero value (compare with Fig. 13). In this regime the system is compensating for the loss of order in the longitudinal direction by building up fluctuations in the transverse

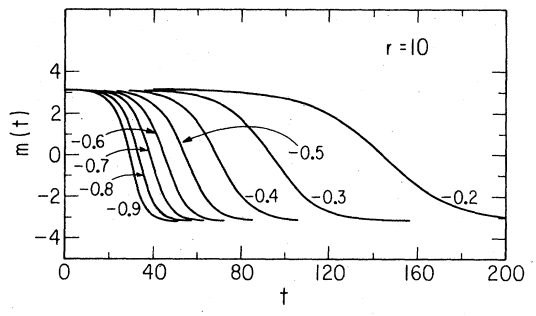

FIG. 11. Time evolution of the magnetization (NCOP) in an isothermal field reversal for various values of h_F and $r_I=-10$.

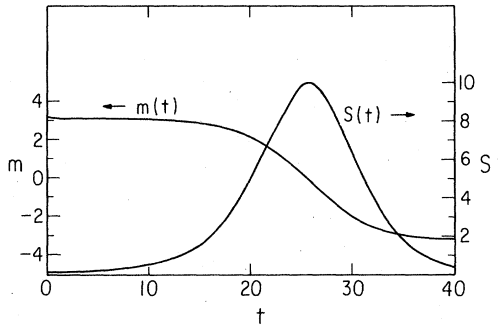

FIG. 13. Time evolution of the magnetization $m(t)$ and of the transverse fluctuations $S(t)$ in the isothermal field reversal. $r=-10$ and $h_F=-1.0$.

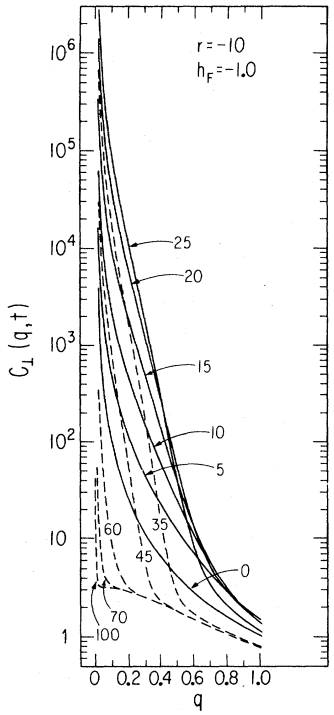

FIG. 14. Time evolution of the transverse structure factor $C_\perp(\mathbf{q},t)$ in the isothermal field reversal for $r=-10$ and $h_F=-1.0$.

directions. However, as the magnetization is flipped, ordering develops in the negative longitudinal direction, and correspondingly the transverse fluctuations are expected to decrease. This is clearly shown by the dashed curves in Fig. 14, which illustrate the relaxation of $C_\perp(q,t)$ to the asymptotic Lorentzian form $C_\perp(q,\infty)=1/(q^2+h_F/m_F)$.

VI. CONCLUSIONS

Our study has led to a clear picture of the growth of order in systems with a continuous symmetry. In the cases of both a COP and a NCOP, we see that the system does it best to build Bragg peaks which characterize the development of order. These contributions satisfy scaling laws similar to those found for scalar order parameters. The growth laws associated with the characteristic length [$L(t)=q_w^{-1}(t)$ for a NCOP and $L(t)=q_M^{-1}(t)$ for a COP] show a power-law behavior $L(t)\sim t^a$, with $a=\frac{1}{2}$ for a NCOP and $a=\frac{1}{4}$ for a COP. The $a=\frac{1}{2}$ result for a NCOP agrees with the $N=1$ result found by Lifshitz, and Cahn and Allen, and suggests that this result may hold for all N for a NCOP. This is not a completely trivial result since the Lifshitz–Cahn-Allen result is built on the assumption of a sharp interface which is not valid for $N>1$. The result $a=\frac{1}{4}$ for a COP may well depend on the large number of degenerate ground states in the model. The standard assumption for the $N=1$ case is that the growth kinetics are given by the Lifshitz and Slyozov theory, which gives $a=\frac{1}{3}$. This theory assumes one has droplets of a minority phase which grow or shrink, depending on their size, via single-particle diffusion. A key assumption is that one has a sharp interface and local equilibrium across that interface. We expect our N vector model to be a highly interconnected structure with Bloch walls separating domains and the deviation from the $\frac{1}{3}$ value should be no surprise. Recently, Mazenko, Valls, and Zhang[16] have found that the growth law for the spin-exchange Kawasaki kinetic Ising model in the absence of a field is of the form $L(t)\sim \ln t$, which again differs from the Lifshitz-Slyozov theory. In this model one finds a percolative morphology with sharp interfaces. The important new ingredient not in the Lifshitz-Slyozov theory or the TDGL model is that diffusion across an interface is activated, and any assumption of local equilibrium at an interface is not justified. In the case of the TDGL model for large N, one expects the nature of the Bloch walls to be such that diffusion is not hindered.

Riding on top of the ordered regions are the Nambu-Goldstone modes which develop over a time scale which must necessarily be slower than that needed to establish domain structures. Thus, the weight in the NG modes grows as $t^{1/2}$ (COP) or t (NCOP), while the weight in the Bragg peak grows as $t^{3/4}$ (COP) and $t^{3/2}$ (NCOP).

In the problem of an isothermal field flip for a NCOP, we find no limit of stability. The magnetization flips after a finite time after the field reversal for all fields flips. The time τ, however, goes as $h^{-1.07}$, which can lead to long quiescent periods before the magnetization flips for small h.

We have chosen to treat the physically relevant case of three spatial dimensions. One could treat the case $d=2$, but the large-N limit is of less physical interest because of the Mermin-Wagner theorem[28] and the role of N-dependent defects in destroying long-range order.

Our calculations here give some insight into calculational schemes[27,29] which were developed to treat the TDGL model for $N=1$. Efforts to go beyond the noninteracting theory typically involved nonsystematic factorization schemes which led to equations very similar to (2.31), but where $\zeta(t)$ was determined in some approximate fashion. As discussed in Refs. 27 and 29, the solutions to these equations tend to long-time solutions where $\zeta(t)\to 0$. Thus one develops an unphysical q^{-2} behavior for the $N=1$ case. One is led to the conclusion that these approximation methods are picking up part of the Nambu-Goldstone mode we found in our analysis here. The conclusion is that the "mean-field theories" with an equation of motion similar to (2.32) are only appropriate to systems with $N>1$ which do not have sharp interfaces.

ACKNOWLEDGMENTS

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