Analytic fixed points of discrete equations of motion: Universality in homogeneous diffusive systems

P. B. Visscher
Department of Physics and Astronomy, University of Alabama, University, Alabama 35486
(Received 24 June 1983; revised manuscript received 9 December 1983)

In this paper a method is presented for analytically calculating the fixed points of scale-coarsening (renormalization) transformations which act on equations of motion for diffusive systems. These fixed points demonstrate the universality of Fick's-law diffusive behavior: All microscopic equations of motion, if they have the correct symmetry, satisfy the correct conservation laws (i.e., particle conservation), and are not too pathological, will look the same on a large space and time scale. Study of the eigenfunctions of the transformation shows that the dominant correction on smaller scales is given by the Burnett equation. The method applied to homogeneous systems in this paper is used in an accompanying paper to obtain new exact results for the universal long-time behavior of inhomogeneous systems in arbitrary dimension.

I. INTRODUCTION

In this paper we will define space- and time-coarsening transformations which act on the space of equations of motion (EOM's) of one-component diffusive systems. The EOM's are parametrized in a discrete form which can be used to describe a very large variety of physical systems (solute molecules diffusing through a solvent, electrons random-walking through a semiconductor, etc.). The discrete EOM describes the system on a distance scale Δr and a time scale Δt; we may describe the system in as great or as little microscopic detail as desired by using small or large Δr and Δt. Applying the space-coarsening transformation S corresponds to viewing the system on a larger space scale $2\Delta r$, and similarly the time-coarsening transformation T yields an EOM on a time scale $2\Delta t$. It will turn out that a combined transformation (essentially T^2S) has a fixed point. That is, there is an EOM which looks the same when you look at it with scales $\Delta r, \Delta t$ or with $2\Delta r, 4\Delta t$. Furthermore, this is an attractive fixed point: An arbitrary system looks more nearly like the fixed point on the scale $2\Delta r, 4\Delta t$ than on the scale $\Delta r, \Delta t$. The fundamental reason for the universality of Fick's law, i.e., its applicability to a wide variety of diffusive systems, is that Fick's law is the fixed-point EOM. These same ideas are applied to disordered systems in an accompanying paper,[1] where they lead to interesting new physical results on long-time tails.

The principal difference between our approach to scale-coarsening and the usual renormalization-group approach to critical phenomena[2] is that we use EOM's (rather than a Hamiltonian description), and that our EOM's are discrete in time as well as in space. This approach allows us to use time-coarsening as well as space-coarsening transformations, which helps us analyze the dynamics of a system. In noncritical systems such as the ones we consider, the statics are trivial, and it is essential to treat the dynamics correctly. There has been considerable work done on the dynamics of critical phenomena by mode-coupling,[3] ϵ-expansion,[4] and position-space renormalization[5,6] techniques. These techniques have used continuous- rather than discrete-time variables, and appear not to allow the exactness possible (at least in diffusive problems) with discrete-time hydrodynamic EOM's.

The purpose of this paper is to derive exact fixed points for a one-component system, and to thereby set up a formalism for discussing fixed points in two-component systems.[1] We also linearize the coarsening transformation around its fixed point; the eigenvalues of this linearization determine how fast various kinds of deviation from Fick's-law behavior (i.e., various eigenvectors) disappear as we increase the scale. By identifying the largest eigenvalues we can predict the corrections to Fick's-law behavior which will be important on a finite distance and time scale.

We can obtain heuristic pictures of the diffusive fixed point and some of the possible corrections (eigenvectors of the coarsening transformation) from Fig. 1. Figure 1(a) shows the probability distribution for the position at time Δt of a particle which started at the origin at time $t=0$ (variously called the diffusive propagator or Green's function). If ρ is a density which obeys Fick's law,

$$\frac{\partial\rho}{\partial t}=D_p\nabla^2\rho\ , \tag{1.1}$$

with diffusivity D_p, this distribution is a Gaussian with a mean-square width $2D_p\,\Delta t$. (The subscript "p" stands for "physical," to distinguish D_p from a dimensionless D to be discussed later.) If we think of Fig. 1(a) as representing the EOM of a diffusive system, we expect this Fick's-law EOM to be a fixed point of a coarsening transformation. To apply a time-coarsening transformation, we need to compute the corresponding distribution for a time scale $2\Delta t$. Assuming that the displacement of the particle in the second interval Δt is independent of that in the first, the new distribution should be a convolution of two of the old ones; this gives a Gaussian with a mean-square width $4D_p\Delta t$, as shown in Fig. 1(b). To make this look more like the original curve, we can perform a space-coarsening

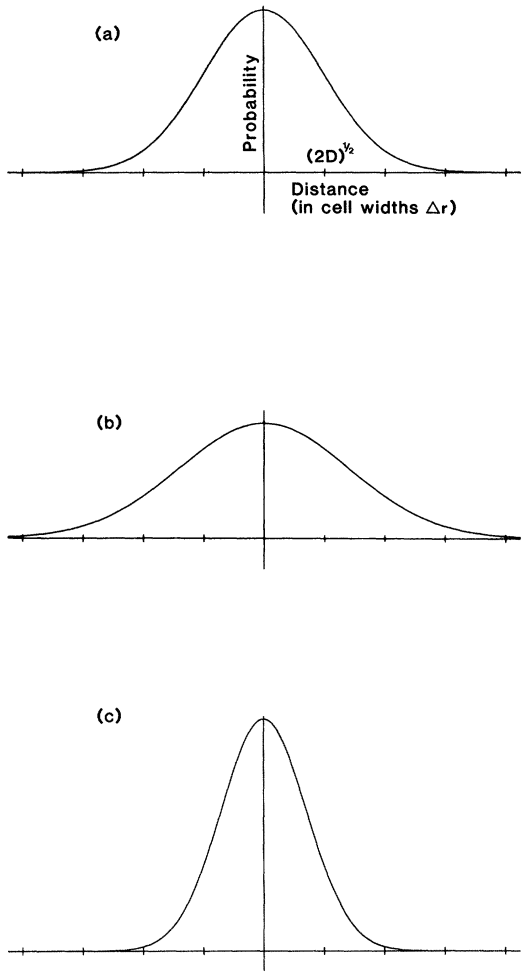

FIG. 1. (a) Propagator for the diffusive fixed point with large dimensionless diffusivity D [proportional to $\exp(-r^2/4D)$, with mean square width $2D$]. (b) After time-coarsening ($D \rightarrow 2D$). (c) After space-coarsening ($2D \rightarrow D/2$).

transformation, replacing the cells of width Δr by cells of width $2\Delta r$. Then the same displacements involve half as many cell widths, i.e., half of the horizontal distance in Fig. 1(b). This shrinks the distribution to that shown in Fig. 1(c), whose mean-square width has decreased by $(\frac{1}{2})^2$ to $D_p t$. Evidently, we must time-coarsen once more (for a total of T^2S, or two time and one space coarsening) to get back to the original width of $2D_p t$ [Fig. 1(a)].

Thus, the EOM represented by Fig. 1(a) is a fixed point[7] of the coarsening transformation T^2S. Clearly, it is not the only one; we could have used any diffusivity D_p and found a fixed point. There is actually a family of fixed points, one for each D_p. To decide whether one of these is a stable fixed point, we must examine whether perturbations around it grow or decay when T^2S is applied. We will do this in Sec. VI by examining the eigenvalues of T^2S linearized about the fixed point; if all of the eigenvalues are less than unity, the fixed point is stable. Here, we examine the eigenvector corresponding to the largest eigenvalue. This eigenvector corresponds to the curve in Fig. 1(a) itself. That is, the perturbation corresponds to adding a small coefficient (for instance, c) multiplied by the Gaussian to the Gaussian. Convoluting the result with itself gives a factor $(1+c)^2$, and T^2S gives $(1+c)^4 \simeq (1+4c)$. Thus the transformation multiplies this perturbation by 4, i.e., the eigenvalue is 4. The Gaussian fixed point is therefore not stable. However, we can stabilize it by requiring our system to satisfy a law of conservation of particles. This restricts the set of EOM's we are considering, but does so in a way that is consistent with the coarsening transformations (both S and T map this subset into itself). The next eigenvector will then turn out to be the derivative with respect to displacement of the Gaussian; it is shown in Fig. 2(a). It has an eigenvalue of 2. Again, the fixed point is unstable until we impose yet another condition on the EOM,, and restrict ourselves to the subset of EOM's which have left-right reflection symmetry. The next eigenvector is the second derivative of the Gaussian [Fig. 2(b)]. Adding an infinitesimal constant c multiplied by this eigenvector to the Gaussian of Fig. 1(a) turns out to give another Gaussian with a diffusivity increased by some δD. Clearly, T^2S gives this same Gaussian back. The perturbation was unchanged, and hence the eigenvector is 1. Thus the fixed point is marginally stable (but clearly a member of a continuous family of fixed points is always marginally unstable in this sense). The first genuinely stable eigenvector is the fourth derivative of the Gaussian. This can be interpreted as giving the system a nonzero Burnett coefficient [coefficient of $\nabla^4\rho$ in the diffusion equation, Eq. (1.1)]. It has an eigenvalue of $\frac{1}{4}$; thus the Burnett effect decreases by a factor of 4 each time we coarsen the scale, and becomes imperceptible on a large enough scale.

In this paper, we will set up a precise and consistent framework for investigating fixed-point properties such as

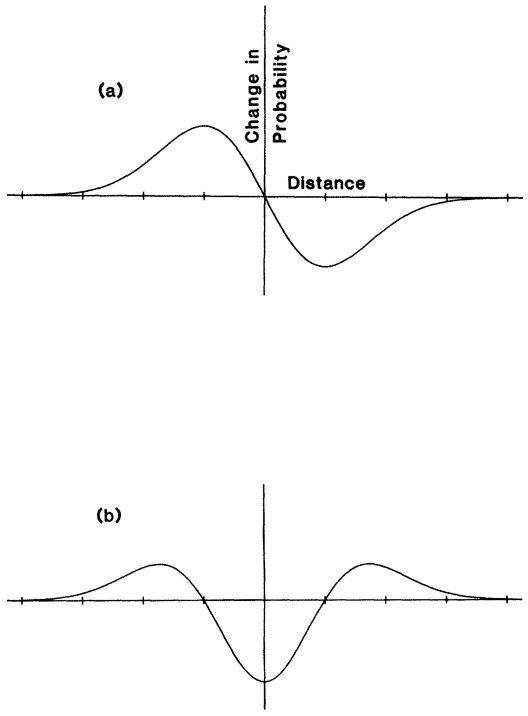

FIG. 2. Eigenvectors of coarsening transformation T^{2S} with (a) eigenvalue 2, and (b) eigenvalue 1.

mentioned above. To do this, we will first review the formulation[8,9] of discrete hydrodynamic EOM's (Sec. II). In Sec. III we describe how to perform space- and time-coarsening transformations on such EOM's. These are Fourier-transformed in Sec. IV. In Sec. V we search for a fixed point and find a two-parameter family of them (Fick's law). We examine the linearized transformation near such a fixed point in Sec. VI, computing eigenvectors and eigenvalues which determine the large-scale behavior of the system. The coefficients of some of the eigenvectors are essentially the physical transport coefficients (Sec. VII).

II. DISCRETE EQUATIONS OF MOTION

Consider a one-component diffusive system. We want to characterize the system by an equation of motion having a specific distance scale Δr and time scale Δt, so that we can perform space- and time-coarsening transformations. The easiest way to do this is to define a set of discrete variables whose evolution will be described by the equation of motion. These variables can be taken to be the contents of cells (d-dimensional cubes of length Δr) at discrete times which are multiples of Δt. We will denote the physical mass content of a cell centered at position r_p at time t_p by $c_p(r_p,t_p)$; this is the total mass of diffusing particles in the cell.[10]

We will want to compare discrete descriptions of the system on different scales (different $\Delta r, \Delta t$) to search for fixed points. The physical variables $c_p(r_p,t_p)$ are not directly comparable: r_p takes on values corresponding to the four solid circles in Fig. 3 on scale Δr, whereas on scale $2\Delta r$ it takes values corresponding to the central $\times$ in the figure. Furthermore, the average content on scale $2\Delta r$ will be larger than on scale Δr. To facilitate such comparisons, let us define a dimensionless position vector $r=r_p/\Delta r$, which points to cell centers in a dimensionless space in which the cells have length 1. We can also adjust for the variable average content by picking a unit of content Δc which is convenient for the particular Δr we are using, e.g., $\Delta c=10^{-15}$ kg if $\Delta r=10^{-6}$ m, and defining a dimensionless mass content

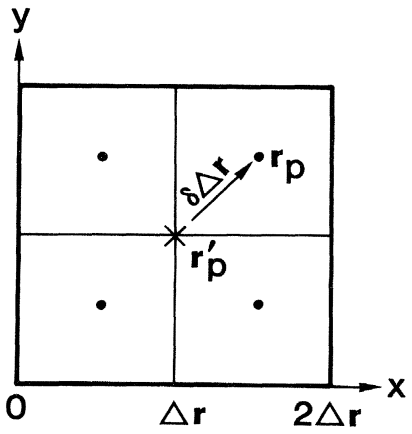

FIG. 3. One coarse cell [at $r'=(\frac{1}{2},\frac{1}{2})$] in $d=2$ dimensions, divided into 2^2 fine cells [$r=(\frac{1}{2},\frac{1}{2})$, $(\frac{1}{2},\frac{3}{2})$, etc.]

$$c(r,t)=(\Delta c)^{-1}c_p(r\,\Delta r, t\,\Delta t)\,, \tag{2.1}$$

where the dimensionless position vector r is the center of a cell (i.e., its components are odd half-integers) and the dimensionless time t is an integer.

Thus for each physical system and each choice of scales Δr, Δt, and Δc, there is a corresponding, unique dimensionless system, with variables $c(r,t)$, which may be directly compared with the dimensionless systems for the same physical system, but with different Δr, Δt, and Δc.

We will analyze the dynamics of a system by examining the dynamics of its dimensionless variables for some particular scales, Δr, Δt, and Δc. The most complete description possible would be the full probability distribution function $P(c)$, which gives the probability of any particular evolution of the system [any choice of $c(r,t)$ for all r,t] in an equilibrium ensemble. Since this is generally difficult,[11] one often considers instead the equilibrium time correlations $\langle c\rangle$, $\langle cc'\rangle$, $\langle cc'c''\rangle$, ..., where the c's can be contents of any cell, at any time. Since this gives all moments of the probability distribution function $P(c)$, it is, in priniciple, equivalent to specifying $P(c)$. The equal-time correlations such as $\langle c(r,0)c(r',0)\rangle$ are often fairly simple, but the unequal-time ones, such as $\langle c(r,t)c(r'0)\rangle$, are typically not. The basic idea of discrete hydrodynamics is that the dynamics of the system can be described more concisely by a set of "equation-of-motion coefficients" which are, in principle, equivalent to the time correlations. First, consider the correlations between $t=0$ and 1. Suppose we knew the conditional moments of $c(r,1)$ in the ensemble whose contents are constrained at $t=0$. The moments will be functions of the values at which the latter are constrained; let us write the moments as power series in these values. Thus the first moment, which we denote by $[c(r,1)]$, is

$$[c(r,1)]=[c(r,1)]_1+\sum_{r'}[c(r,1)]_{c(r',0)}c(r',0) + \sum_{r',r''}[c(r,1)]_{c(r',0)c(r'',0)}c(r',0)c(r'',0) + \cdots\,, \tag{2.2}$$

where each subscripted $[c(r,1)]$ is a power-series coefficient describing the dependence of $c(r,1)$ on the variable(s) in the subscript (the subscript 1 indicates the coefficient of 1, i.e., the constant term.) By subtracting a constant from each c, we can redefine the variables so that $\langle c(r,t)\rangle=0$; this leads to $[c(r,1)]_1=0$, and we will omit such constant terms below. Thus the leading term remaining in Eq. (2.2) involves $[c(r,1)]_{c(r',0)}$, which we can think of as a propagator describing the influence of the content $c(r',0)$ on the later content $c(r,1)$.

The knowledge of all these equation-of-motion coefficients (together with the equal-time correlations) is equivalent to knowing all the equilibrium correlations involving the two times 0 and 1. We will not prove this rigorously, but note that one can write relations between them by multiplying each equation similar to (2.2) by a product of $c(r,0)$'s and averaging over the equilibrium ensemble. In the case of Eq. (2.2), multiplying by $c(r'',0)$ gives

$$\langle c(r,1)c(r'',0)\rangle = \sum_{r'} [c(r,1)]_{c(r',0)} \langle c(r',0)c(r'',0)\rangle + \cdots . \tag{2.3}$$

If we know either the time correlations on the left-hand side or the EOM coefficients on the right-hand side, we can solve for the other. As yet, we have gained very little advantage over using the unequal-time averages; the propagator $[c(r,1)]_{c(r',0)}$ is not actually simpler than the time-correlation $\langle c(r,1)c(r',0)\rangle$ (indeed, at the fixed point they are essentially the same). However, describing the system completely required including correlations involving more times than just 0 and 1. The advantage of the present scheme is that we can do this by just adding more EOM coefficients, such as those in Eq. (2.2), but involving ensembles constrained at successively more times. If we call the number of constrained times m, then the equal-time equilibrium averages such as $\langle c(r,1)c(r',1)\rangle$ correspond to the case $m=0$ (nothing constrained), and the EOM coefficients in Eq. (2.2) to $m=1$ (one time, $t=0$, constrained). In general, we define the mth set of EOM coefficients to describe an ensemble in which contents at m times ($1-m$ through 0) are constrained. If we think of the "zeroth set of EOM coefficients" as being the equilibrium averages, what we showed above is that the zeroth and first sets together determine all time correlations among the two times 0 and 1. It can be shown in an exactly similar way[9] that the zeroth, first, . . . , mth sets together determine all time correlations among the $m+1$ times $1-m \le t \le 1$. The collection of *all* such EOM coefficients (for all m) therefore uniquely determines the system.

The above parametrization of the EOM has been used for numerical calculations on fluid systems which are non-Markovian in the sense that several m's are needed.[9] However, our present interest is in Markovian systems in the small-cell ($\Delta r \to 0$) limit, for which $m=0$ and 1 are entirely sufficient. This is because the second set of coefficients, having $m=2$, describes the effects of $c(r,-1)$ on the future content $c(r,1)$, as well as the effects of $c(r,0)$. However, the sufficiency of $c(r,0)$ to determine the future evolution is, of course, what is meant by "Markovian" [at least in the continuum limit $\Delta r \to 0$; $c(r,-1)$ is necessary if Δr is large[8]]. Therefore, we may regard our system as being uniquely determined by the equal-time correlations and the $m=1$ EOM coefficients of Eq. (2.2). Furthermore, it will turn out that the fixed-point EOM's and the most important perturbations around them are both linear and Gaussian. By the latter, we mean that the equal-time fluctuations are Gaussian, and thus the second moment

$$\langle c(r,0)c(r',0)\rangle \tag{2.4a}$$

uniquely determines the equilibrium correlations. In a linear system, the linear propagator

$$[c(r,1)]_{c(r',0)} \tag{2.4b}$$

is the only EOM coefficient we need to retain in Eq. (2.2). Our system is therefore specified by only the two quantities (2.4a) and (2.4b).

III. COARSENING TRANSFORMATIONS

Let us first define the space-coarsening transformation S, which acts on dimensionless discrete equations of motion. Consider a dimensionless EOM E giving the dynamics of dimensionless variables $c(r,t)$, which describes a physical system on the scale $\Delta r, \Delta t, \Delta c$ with physical variables $c_p(r_p,t_p)$, through Eq. (2.1),

$$c_p(r_p,t_p) = (\Delta c)c(r_p/\Delta r, t_p/\Delta t) . \tag{3.1}$$

We define the space-coarsened EOM, denoted SE, as the dimensionless equation of motion which describes this same physical system on the coarser scale $2\Delta r, \Delta t, \Delta c$. Thus the dimensionless variables $c'(r',t')$ of SE (we will indicate coarse-scale quantities by primes) are related to large-cell physical variables,

$$c'_p(r'_p,t'_p) = (\Delta c)c'(r'_p/2\Delta r, t'_p/\Delta t) . \tag{3.2}$$

To see that this uniquely defines SE for each E let us calculate a particular description of SE explicitly. It is easiest to describe SE by the equilibrium moments of the contents, so that SE is specified by equilibrium averages such as

$$\langle c'(r'_1,t'_1)c'(r'_2,t'_2)\cdots c'(r'_n,t'_n)\rangle . \tag{3.3}$$

In terms of the physical variables [Eq. (3.1)] this is

$$(\Delta c)^{-n}\langle c'_p(2r'_1\Delta r, t'_1\Delta t)\cdots c'_p(2r'_n\Delta r, t'_n\Delta t)\rangle . \tag{3.4}$$

The coarse cell at $r'_p = 2r'\Delta r$ is made up of exactly 2^d smaller ("fine") cells (see Fig. 3). Its content c'_p is exactly the sum of 2^d fine contents c_p,

$$c'_p(2r'\Delta r, t'\Delta t) = \sum_{\delta} c_p((2r'+\delta)\Delta r, t'\Delta t) , \tag{3.5}$$

where each component of the dimensionless displacement vector δ is $\pm\frac{1}{2}$. The coarse moment [expression (3.3)] is then expressible in terms of fine moments; it is

$$\begin{aligned}\langle c'(r'_1,t'_1)\cdots\rangle &= (\Delta c)^n \sum_{\delta_1}\cdots\sum_{\delta_n}\langle c_p((2r'_1+\delta_1)\Delta r, t'_1\Delta t)\cdots c_p((2r'_n+\delta_n)\Delta r, t'_n\Delta t)\rangle \\ &= \sum_{\delta_1}\cdots\sum_{\delta_n}\langle c(2r'_1+\delta_1,t'_1)\cdots c(2r'_n+\delta_n,t'_n)\rangle\end{aligned} \tag{3.6}$$

[we have again used Eq. (3.1)]. Thus we can compute SE directly from E by merely adding averages, and the space-coarsening transformation is well defined. It is not so trivial to compute SE when E is specified by equation-of-motion coefficients as in Sec. II, but a procedure for doing so has been described and numerically implemented.[8]

Let us now define the time-coarsening transformation

T. We can define TE as the dimensionless EOM which describes our same physical system on the scales $\Delta r, 2\Delta t, \Delta c$. Thus it should describe the dynamics of physical variables $c'_p(r_p, t'_p)$ which are exactly those of E, except that half of them (those with odd t) are left out (see Fig. 4). Unlike S, which involved a lumping together of fine variables, T simply involves a decimation process, that is, the elimination of some of the variables. Explicitly,

$$\langle c'(r'_1,t'_1)\cdots c'(r'_n,t'_n)\rangle = \langle c'(r'_1,2t'_1)\cdots c'(r'_n,2t'_n)\rangle \tag{3.7}$$

gives TE in terms of E. An algorithm for applying T to an equation of motion has also been described.[8]

In addition to the coarsening transformations S and T, we can also define a content rescaling transformation we will denote by R. It is defined analogously to S and T: If E describes a system with scales $\Delta r, \Delta t, \Delta c$, then RE describes the same system with scales $\Delta r, \Delta t, 2\Delta c$. This induces a trivial transformation on the averages, $c'(r,t) = \frac{1}{2}c(r,t)$, and so

$$\langle c'(r_1,t_1)\cdots c'(r_n,t_n)\rangle = 2^{-n}\langle c(r_1,t_n)\cdots c(r_n,t_n)\rangle \ . \tag{3.8}$$

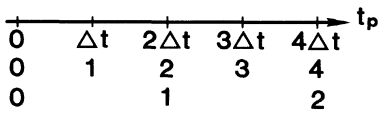

FIG. 4. Relation between the physical time t_p and the fine (t) and coarse (t') dimensionless time variables.

The R transformation is nevertheless important because combinations of S and T alone have no fixed points.

IV. FOURIER TRANSFORMATION OF DISCRETE EQUATIONS OF MOTION

In this paper we will deal with the limit in which the cell size Δr is very small compared to the characteristic lengths of the problem, such as $(D_p \Delta t)^{1/2}$. In this continuous-space limit the EOM coefficients and averages are smoothly varying and it makes sense to deal with them in k space.

Consider an EOM coefficient

$$[c(r_1,t_1)\cdots c(r_s,t_s)]_{c(r_{s+1},t_{s+1})\cdots c(r_n,t_n)} \ ,$$

as in Sec. II, having s predicted and $n-s$ constrained variables. We will define its Fourier transform G^s_{n-s} by

$$(2\pi)^d \delta_{2\pi}(k_1+\cdots+k_s-k_{s+1}-\cdots-k_n) G^s_{n-s}(k_1,t_1,\ldots,k_s,t_s;k_{s+1},t_{s+1},\ldots,k_n,t_n)$$
$$= \sum_{r_1}\cdots\sum_{r_n} \exp(-ik_1r_1\cdots-ik_sr_s+ik_{s+1}r_{s+1}+\cdots+ik_nr_n)[c(r_1,t_1)\cdots c(r_s,t_s)]_{c(r_{s+1},t_{s+1})\cdots c(r_n,t_n)} \ . \tag{4.1}$$

The Dirac δ function has been factored out because the translational periodicity of the system makes the right-hand side singular; it is zero unless the sum $k_1+\cdots-k_n$ is zero (mod 2π). G need to only be defined for such k's. The function $\delta_{2\pi}$ is a d-dimensional periodic δ function, defined by

$$\delta_{2\pi}(k) = \sum_p \delta(k+2\pi p) \ ,$$

where the p's are d-tuples of integers.

Evidently, G has a periodicity property in each k_i: If the bth spacial component of the vector k_i is replaced by $k_i^b + 2\pi$, G changes by a factor $\exp(\pm 2\pi i r_i^b)$. Because each r_i^b is a half-integer, this factor is -1: G is antiperiodic in each k_i^b, with period 2π. The inversion formula is

$$[c(r_1,t_1)\cdots]_{c(r_{s+1},t_{s+1})\cdots} = \int \frac{dk_1}{(2\pi)^d} e^{ik_1r_1}\cdots$$
$$\times \int \frac{dk_{s+1}}{(2\pi)^d} e^{-ik_{s+1}r_{s+1}}\cdots(2\pi)^d\delta_{2\pi}(k_1+\cdots k_s-k_{s+1}-\cdots-k_n)\cdots$$
$$\times G^s_{n-s}(k_1,t_1,\ldots,k_s,t_s;k_{s+1},t_{s+1},\ldots,k_n,t_n) \ . \tag{4.2}$$

The integrals are over hypercubes, 2π on each side, which can be located anywhere because of the antiperiodicity in each k_i. The inversion formula can be verified using either of the identities

$$\int \frac{dk}{(2\pi)^d} e^{ik(r-r')} = \delta_{rr'} \tag{4.3}$$

or

$$\sum_r e^{i(k-k')(r-1/2)} = (2\pi)^d \delta_{2\pi}(k-k') \tag{4.4}$$

(here $\frac{1}{2}$ denotes a vector each of whose components is $\frac{1}{2}$).

In the case of $n=2$ variables of which $s=2$ are predicted, Eqs. (4.1) and (4.2) define a Fourier transform G^2_0 of the conditional fluctuation $[c(r_1,1)c(r_2,1)]$. Since we are describing fluctuations in terms of the equilibrium average $\langle c(r_1,0)c(r_2,0)\rangle$ instead, we may use the same equations to compute *its* Fourier transform, which we denote by $G^2_{\rm eq}(k_1,0,k_2,0)$.

In terms of the G's, the quantities that we concluded in Sec. II were sufficient to describe a linear, Gaussian, Mar-

kovian system are $G_1^1(k,1,k,0)$ and $G_{eq}^2(k,0,-k,0)$. Since the last three arguments are redundant, we will omit them and denote the propagator by $G_1^1(k)$ and the equilibrium average by $G_{eq}^2(k)$.

We must now examine the problem of carrying out coarsening transformations in the Fourier representation. First, considering space-coarsening, it is shown in the Appendix that the coarsened $G_{eq}^{2\prime}$, and $G_1^{1\prime}$, are given by

$$G_{eq}^{2\prime}(k')=2^d G_{eq}^2(k'/2) , \tag{4.5a}$$

$$G_1^{1\prime}(k')=G_1^1(k'/2) . \tag{4.5b}$$

The rescaling $k'\to k'/2$ is certainly to be expected, and 2^d appears because it is the volume scaling factor, the number of fine-cell contents we add to obtain one coarse-cell content.

Now consider time-coarsening in the Fourier representation. We defined the time-coarsening transformation T in terms of equilibrium correlations [Eq. (3.7)]. Coarsening the equal-time average is then trivial: It is independent of Δt, and

$$G_{eq}^{2\prime}(k)=G_{eq}^2(k) , \tag{4.6a}$$

as may be formally verified from Eqs. (3.7) and (4.1). We are describing the unequal-time properties by the propagator G_1^1, which cannot be time-coarsened so directly. A very general graphical procedure for coarsening it is given in the following paper,[1] but the result we require here can be seen more easily by thinking of the propagator as describing the distribution of positions at $t=1$ of a particle which began at the origin at $t=0$ (Fig. 1). Evidently, the distribution after two intervals is just the convolution with another identical propagator (in real space), or in k space,

$$G_1^{1\prime}(k)=[G_1^1(k)]^2 . \tag{4.6b}$$

Equations (4.6) are the only ones we will need here for time-coarsening.

For completeness we should also write the equations for applying the rescaling transformations R to an EOM. From Eq. (3.8),

$$G_{eq}^{2\prime}(k)=2^{-2}G_{eq}^2(k) , \tag{4.7a}$$

$$G_1^{1\prime}(k)=G_1^1(k) . \tag{4.7b}$$

V. FIXED POINTS

We now have formulas for space-coarsening [Eqs. (4.5a) and (4.5b)], time-coarsening [Eqs. (4.6a) and (4.6b)], and rescaling [Eqs. (4.7a) and (4.7b)], a discrete equation of motion E, which is described by two functions G_{eq}^2 and G_1^1 of a single vector variable k in the limit we are interested in (which we may call the continuous-space or Markovian limit). It is evident that no EOM can be fixed under S [which continually spreads out the propagator G_1^1 in k space via Eq. (4.5b)] or T [which continually spreads G_1^1 out in real space by convoluting it; Eq. (4.6b)]. Thus, we seek a fixed point under some unknown combination ST^zR^y of the transformations. First examining the equal-time average G_{eq}^2, S spreads it out in k space, and neither T nor R affects its range. If we assume on physical grounds that G_{eq}^2 starts out finite-ranged in space (nonzero-ranged in k), any fixed point it approaches must be zero-ranged in space. This is infinite-ranged in k, i.e., a constant

$$G_{eq}^2(k,0;-k,0)=A . \tag{5.1}$$

This corresponds in real space [using Eq. (4.2)] to

$$\langle c(r,0)c(r',0)\rangle = A\,\delta_{rr'} \tag{5.2}$$

($\delta_{rr'}$ is the Kronecker δ), and thus A is the mean-square dimensionless content.

Let us check that Eq. (5.1) is actually fixed under ST^zR^y. From Eq. (4.5a), S turns A into 2^dA; schematically S: $A\to 2^dA$. The time-coarsening transformation T has, of course, no effect on the single-time average G_{eq}^2, but Eq. (4.7a) implies $R:A\to 2^{-2}A$. Thus

$$ST^zR^y:A\to 2^d2^{-2y}A ,$$

and we have a fixed point if

$$y=d/2 . \tag{5.3}$$

We now need to find a propagator G_1^1 which is fixed. Denoting it temporarily by G,

$$G(k)=G_1^1(k,1;k,0) , \tag{5.4}$$

we obtain, from Eq. (4.5b), S: $G(k)\to G(k/2)$. Since R has no effect and T: $G(k)\to G(k)^2$ [Eq. (4.6b)], we have $ST^zR^{d/2}$: $G\to G'$, where

$$G'(k')=G(k'/2)^{(2^z)} . \tag{5.5}$$

Requiring $G'=G$ and taking logarithms,

$$\ln G(k)=2^z\ln G(k/2) . \tag{5.6}$$

The solution for $\ln G$ is then evidently a homogeneous function of k, whose order is z: $\ln G=-Dk^z$ for some constant $-D$. Thus,

$$G(k)=\exp(-Dk^z) . \tag{5.7}$$

We can now look at possible values of z; if we require G to be analytic it is a non-negative integer. The value $z=0$ is forbidden if we require conservation of mass,

$$1=\sum_{r'}[c(r,1)]_{c(r',0)}=G(k=0) . \tag{5.8}$$

Inversion symmetry [$G(k)=G(-k)$] forbids odd z. Thus we are left with $z=2,4,6,\ldots$. Each of these gives a fixed point under the corresponding transformation $ST^zR^{d/2}$. The ones for $z=2$ and 4 have previously been calculated numerically (Refs. 8 and 12). It will turn out that only $z=2$ gives a stable fixed point, so we will concentrate on it here. It evidently corresponds to Fick's law of diffusion; we will call it a Fick's-law fixed point. The constant D is then a dimensionless diffusivity [the physical diffusivity will be shown in Sec. VIII to be $D(\Delta r)^2/\Delta t$)].

In summary, we have constructed a family of fixed points under $ST^2R^{d/2}$ which is parametrized by the dimensionless diffusivity D and the mean-square content

fluctuation A (recall that we subtracted a constant so that the mean content vanished). We may denote the equation of motion determined by D and A by $E(A,D)$; it is defined only for $D \gg 1$, by Eqs. (5.1) and (5.8).

VI. EIGENFUNCTIONS AND EIGENVALUES: UNIVERSAL LARGE-SCALE BEHAVIOR

A particular diffusive physical system will have an equation of motion which approaches one of the fixed points computed in the preceding section as it is repeatedly coarsened. The results of very-large-scale (in space and time) measurements will therefore depend only on the fixed point, i.e., on D and A. Somewhat smaller-scale measurements, however, may provide information about the EOM just before it reaches the fixed point. The effects of the coarsening transformation $ST^2R^{d/2}$ in the immediate vicinity of the fixed point can be described in terms of the linearized transformation L about the fixed point, defined as follows: If E^* is the fixed point $E(A,D)$, and $E^*+\delta E$ is a nearby EOM,

$$L\,\delta E \equiv (ST^2R^{d/2})(E^*+\delta E)-E^* .$$

(We may define the operation of subtraction on EOM's by subtracting the functions $G^2_{\rm eq}$ and G^1_1 which define them; in the linear region around E^* this should be equivalent to subtracting equilibrium averages.) If we can find a complete set of eigenfunctions E_i and eigenvalues μ_i of L, we can expand

$$\delta E = \sum_i \lambda_i E_i \tag{6.1}$$

and predict its further trajectory under coarsening: $ST^2R^{d/2}$: $\lambda_i \to \mu_i \lambda_i$. The large-scale behavior of the system will be determined by the eigenfunctions whose coefficients decay most slowly as we coarsen the scale, i.e., those with the largest eigenvalues μ_i. Even if we cannot find a complete set, it is useful for our purposes to identify these largest eigenvalues.

A very general but somewhat abstract method for finding eigenfunctions is given in the accompanying paper.[1] Here we will use a more direct approach. We want to find a perturbation in the EOM, i.e., small changes in the functions $G^2_{\rm eq}$ and G^1_1 which describe it, which corresponds to an eigenfunction. We will begin by letting the equal-time average $G^2_{\rm eq}$ stay the same, and perturbing the propagator $G(k)=G^1_1(k,1,k,0)$ from its fixed-point value [Eq. (5.7)], $G^*(k)=\exp(-Dk^2)$, to $G^*+\delta G$. The eigenvalue equation is

$$ST^2R^{d/2}(G^*+\delta G)=G^*+\mu\,\delta G \tag{6.2}$$

or [using Eqs. (4.5b) and (4.6b)]

$$[G^*(k/2)+\delta G(k/2)]^4 = G^*(k)+\mu\,\delta G(k) .$$

Linearizing in δG gives

$$4\frac{\delta G(k/2)}{G^*(k/2)} = \mu \frac{\delta G(k)}{G^*(k)} . \tag{6.3}$$

This has the same form as Eq. (5.6), and the same homogeneous-function solution,

$$\frac{\delta G(k)}{G^*(k)} = \lambda k^j , \tag{6.4}$$

which corresponds to the eigenvalue

$$\mu = 2^{2-j} . \tag{6.5}$$

This gives an analytic function of k for any integer $j \geq 0$, and thus we have found a series of eigenfunctions which we can label by these j's, and which turn out to be exactly those we described heuristically in Sec. I. To interpret them, examine

$$G(k)=G^*+\delta G=\exp(-Dk^2)(1+\lambda k^j) ,$$

which is the same to linear order as

$$G(k)=\exp(-Dk^2+\lambda k^j) . \tag{6.6}$$

Evidently, $j=0$ causes $G(k)$ to be multiplied by a constant e^λ. This violates conservation of mass [Eq. (5.8)], but is mathematically a perfectly good eigenvalue. The $j=1$ eigenfunction was shown in Fig. 2(a), but odd j violates inversion symmetry. The next even value, $j=2$ [Fig. 2(b)], simply changes the value of D and thus has an eigenvalue $\mu=1$, as mentioned in Sec. I.

Note that we can write a propagator perturbed by a linear combination of these eigenfunctions in the form

$$G(k)=\exp(-D_0-D_2k^2-D_4k^4-\cdots) \tag{6.7}$$

(leaving out odd j for brevity), which is useful because it makes sense beyond first order in the D_j's. Thus D_0 describes the particle decay rate (degree of nonconservation), and D_2 is the dimensionless diffusion coefficient that we denoted D previously. In terms of this notation, the largest allowed eigenvalue corresponding to a deviation from the family of fixed points (i.e., from Fick's law of diffusion) is $\mu=\frac{1}{4}$. The corresponding amplitude D_4 is the dimensionless Burnett coefficient.[13,14]

We must also consider perturbations of the equal-time average $G^2_{\rm eq}(k,0,-k,0)$, which we may denote by $G_{\rm eq}(k)$. The fixed-point value is $G^*_{\rm eq}(k)=A$. Arguments similar to the above lead to eigenvectors k^i $(i \geq 0)$ and eigenvalues $\mu_i = 2^{-2i}$. Here i must be even since $\langle c(r,0)c(r',0)\rangle$ is unchanged when $r-r'$ changes sign. The case $i=0$ corresponds to moving along the family of fixed points, and has $\mu=1$. The perturbed $G_{\rm eq}$ can be written

$$G_{\rm eq}(k)=A\exp(-\xi^2k^2-\lambda_4k^4) . \tag{6.8}$$

We have written the amplitude for $i=2$ as ξ^2 instead of λ_2, since this is the standard notation for a correlation length. This correlation-length eigenvector has eigenvalue $\mu=\frac{1}{4}$, the same as the Burnett eigenvalue. In principle, we should also consider perturbation of the average content $\langle c(r,0)\rangle$, which is uniform in space, or its Fourier transform $G^1_{\rm eq}(k,0)$. This is easily seen to grow with eigenvalue $2^{d/2}$; it is highly unstable. That is why it was necessary to subtract off the average content after Eq. (2.2).

We now know various eigenfunctions and eigenvalues of the coarsening transformation $ST^2R^{d/2}$. Just as in the theory of critical phenomena, this knowledge enables us to

predict the universal large-scale behavior of a diffusive system. As we examine larger scales (i.e., apply $ST^2R^{d/2}$), the perturbations from Fick's law corresponding to stable eigenfunctions ($\mu<1$) will become smaller (we have constrained the amplitudes of eigenfunctions with $\mu\geq 1$ to vanish on all scales). The EOM will therefore approach the Fick's-law fixed point. The eigenfunctions which disappear most slowly, i.e., the ones that are most visible on a relatively large scale, will be those with the largest eigenvalues. We have not proved that the stable eigenvalues we have found are the largest (indeed, there are larger ones if we allow nonlinear EOM's, as we see in the following paper on disordered systems[1]). However, they appear to be the largest ones, if we restrict ourselves to linear systems. There are two eigenfunctions with the same eigenvalue $\mu=\frac{1}{4}$, corresponding to a nonzero Burnett coefficient and a nonzero correlation length, respectively. These determine the leading correction to the dynamic structure function $S(k,\omega)$ obtained from Fick's law, for example.

VII. PHYSICAL TRANSPORT COEFFICIENTS

In terms of a continuum equation of motion such as Eq. (1.1), what we have done, in effect, is to confirm the Burnett generalization

$$\frac{\partial\rho}{\partial t}=-D_{0p}\rho+D_{2p}\nabla^2\rho-D_{4p}\nabla^4\rho+\cdots\ , \tag{7.1}$$

where D_{4p} is the physical Burnett coefficient; we have also included the decay rate D_{0p} for generality. (The signs must be chosen this way, with positive D's for physical stability.) Let us demonstrate explicitly the connection between the dimensionless coefficients D_j of our eigenvectors [as in Eq. (6.7)] and the physical transport coefficients D_{jp} in Eq. (7.1). First, we Fourier-transform Eq. (7.1),

$$\frac{\partial\rho}{\partial t}=-D_{0p}\rho-D_{2p}k_p^2\rho-D_{4p}k_p^4\rho-\cdots \tag{7.2}$$

(here $k_p=k/\Delta r$), and solve it with initial condition $\rho(k_p,0)$,

$$\frac{\rho(k_p,t)}{\rho(k_p,0)}=\exp(-D_{0p}\Delta t-D_{2p}k_p^2\Delta t-\cdots)\ . \tag{7.3}$$

Letting $\rho(k_p,0)$ be independent of k_p (an initial distribution concentrated at the origin), we can identify the ratio $\rho(k_p,t)/\rho(k_p,0)$ with the propagator $G(k)$. Comparison with Eq. (6.7) gives

$$D_{jp}=D_j(\Delta r)^j/\Delta t\ . \tag{7.4}$$

In particular, the physical Burnett coefficient is

$$D_{4p}=D_4(\Delta r)^4/\Delta t\ .$$

Note that under coarsening this changes by $\frac{1}{4}(2)^4/2^2=1$, i.e., the physical Burnett coefficient is invariant. This is true of all the D_{jp}'s.

VIII. SUMMARY

We have shown that a general understanding of the large-scale behavior of diffusive systems can be obtained by analyzing the exact eigenfunctions and eigenvalues of a linearized coarsening transformation near a family of fixed-point equations of motion. The fixed points are parametrized by two dimensionless parameters A and D_2 related to the content-fluctuation amplitude and the diffusivity. The coefficients of the two most relevant eigenfunctions are related to the correlation length and the Burnett coefficient; this means that these are the dominant corrections to normal (Fick's-law) diffusive behavior. In the following paper these techniques are generalized to a two-component system, and are used to obtain new results for the universal behavior of systems with static disorder.

ACKNOWLEDGMENT

This work was supported by the Chemistry Division of the National Science Foundation under Grant No. CHE-8106122.

APPENDIX

In this appendix we give a general derivation of a formula for space-coarsening a Fourier-transformed EOM, which leads to Eqs. (4.5) in a certain Markovian limit.

In previous work we have space-coarsened EOM's numerically[8,12] using a procedure which used the EOM propagators directly. In the present case (a Markovian EOM in the limit $\Delta r\to 0$) there is a simple relation [Eq. (2.3)] between the propagator G_1^1 and the equilibrium averages, which we can Fourier-transform [via Eqs. (4.1)—(4.4)] to

$$G_{\rm eq}^2(k,1,-k,0)=G_1^1(k,1,k,0)G_{\rm eq}^2(k,0,-k,0)\ . \tag{A1}$$

Thus it is sufficient to S-coarsen the equilibrium time correlations; we can then obtain the propagator from Eq. (A1). The equation for the S-coarsened time correlation is simple in real space [Eq. (3.6)]. It is also simple in k space [Eqs. (4.5), derived below] in the limit where $\Delta r\to 0$. To show this and estimate the errors for $\Delta r\neq 0$, however, we need to Fourier-transform Eq. (3.6) for nonzero Δr. Substituting it into Eq. (4.1), which defines the Fourier transform, gives an expression for the coarsened $G_{\rm eq}^{n'}(k_1',t_1,\ldots,k_n',t_n)$ in terms of $G_{\rm eq}^n$. The expression contains sums over cell coordinates

$$\sum_{r'}e^{-ik'r'}e^{ik(2r')}\ .$$

Considering only a single component of k', k, and r', this can be replaced [using Eq. (4.4)] by

$$2\pi\,\delta_{2\pi}(k'-2k)e^{i(k'-2k)/2}=\pi\,\delta_\pi(k'/2-k)e^{i(k'/2-k)}\ ,$$

where δ_π has period π. It can, however, be expressed as the sum of two $\delta_{2\pi}$'s,

$$2\pi\delta_{2\pi}(k'-2k)e^{i(k'-2k)/2}=\pi\sum_\theta\delta_{2\pi}(k'/2-k+\theta)e^{i\theta}\ ,$$

where θ takes on the two values 0 and π. Thus the sum

over vector positions r' is

$$\sum e^{-ik'r'}e^{ik(2r')}=\pi^d\sum_\theta \delta_{2\pi}(k'/2-k+\theta)e^{i\sum\theta}\ ,$$

where θ is now a vector with components 0 or π, and $\sum\theta$ is the sum of the components. This δ function eliminates the k integrals in Eq. (4.1), leaving

$$\delta_{2\pi}(k'_1+\cdots)G^{n'}_{\rm eq}(k'_1,\ldots)=\sum_{\delta_1}\sum_{\theta_1}2^{-d}\exp\left[i\sum\theta_1+i(k_1/2+\theta_1)\delta_1\right]$$
$$\times\sum_{\delta_2}\sum_{\theta_2}\cdots\delta_{2\pi}(k'_1/2+\theta_1+k'_2/2+\theta_2+\cdots)G^{n}_{\rm eq}(k'_1/2+\theta_1,\ldots)\ . \qquad \text{(A2)}$$

This is nonzero only when each component of $\sum k'_1$ is a multiple of 2π. When it is an even multiple, the δ function on the right-hand side requires each component of $\sum\theta_i$ to be an even multiple of π, and

$$\delta(k'_1/2+\theta_1+\cdots)=\delta(k'_1/2+k'_2/2+\cdots)=2^d\delta(k'_1+k'_2+\cdots)\ .$$

Thus Eq. (A2) simplifies to

$$G^{n'}_{\rm eq}(k_1,\ldots)=2^d\sum_{\theta_1}\cdots\sum_{\theta_n}\cos(k'_1/4+\theta_1/2)\cdots\cos(k'_n/4+\theta_n/2)G^{n}_{\rm eq}(k'_1/2+\theta_1,\ldots)\ , \qquad \text{(A3)}$$

where the sum is over $\theta_i=0,\pi$, and includes only θ's for which each component of $\sum\theta_i/\pi$ is even. We are using the notation $\cos(k)$ for a vector $k=(k^1,k^2,\ldots,k^d)$ to mean $\cos(k^1)\cos(k^2)\cdots\cos(k^d)$. Because of the antiperiodicity there is no need to evaluate $G^n_{\rm eq}$ when a component of $\sum k'_1/2\pi$ is odd; however, Eq. (A3) works for this case provided we require the corresponding component of $\sum\theta_i/\pi$ to be odd and insert a factor -1.

It is difficult to find analytic expressions for $G^n_{\rm eq}$ which are fixed points of a transformation such as Eq. (A3) involving a sum over displacements θ. In our calculations we will assume that G is strongly peaked near the origin, falling off, for example, as $\exp(-\xi^2k_i^2)$ in each of its arguments k_i, with a correlation length $\xi\gg1$. Of course, there is another peak at $k_i=2\pi$, but the peaks are separated by a region in which the value is very small, of order $\exp(-\xi^2\pi^2)$, and because of the antiperiodicity they are all determined by the one near $k_i=0$. When we evaluate Eq. (A3) near $k_i=0$, the terms with $\theta_i\neq0$ will be negligible, and the space-coarsening equation simplifies to

$$G^{n'}_{\rm eq}(k'_1,t_1,\ldots)=2^d\cos(k'_1/4)\cdots\cos(k'_n/4)G^{n}_{\rm eq}(k'_1/2,t_1,\ldots)\ , \qquad \text{(A4)}$$

with an error of order $\exp(-\xi^2\pi^2)$.

For the present purposes, we shall also make a much more severe approximation by expanding the cosines in powers of k_i and keeping only the constant term,

$$G^{n'}_{\rm eq}(k'_1,t_1,\ldots)=2^dG^{n}_{\rm eq}(k'_1/2,t_1,\ldots)\ . \qquad \text{(A5)}$$

The error introduced by this is of relative order ξ^{-2}.

Equation (A5) can be used to space-coarsen the equilibrium averages $G^2_{\rm eq}$ in Eq. (A1), and therefore to also determine the coarsened propagator $G^{1'}_1$, which is given by Eq. (4.5). Equation(4.5) is therefore valid for a Markovian EOM in the continuum ($\Delta r\to0$) limit as required in Sec. IV for large correlation length ξ.

The reader may note, however, that the fixed point we finally end up with [Eq. (5.1)] violates the assumption $\xi\gg1$; in fact, $\xi=0$. Indeed, the argument for Eq. (5.1) amounts to observing that S: $\xi\to\xi/2$ (S changes ξ to $\xi/2$), T: $\xi\to\xi$, and R: $\xi\to\xi$ so $\xi=0$ at any fixed point. (Remember that ξ is a dimensionless correlation length, i.e., the number of cell lengths across which the density is correlated, which decreases as the cells enlarge.) Fortunately, Eq. (4.5a) can still be proved in this special case. Applying the exact Eq. (A3) to Eq. (5.1) and noting that the evenness condition on θ_i can be satisfied by taking $\theta_2=-\theta_1$, we obtain

$$G^{2'}_{\rm eq}(k',0,-k',0)=2^d\sum_{\theta=0}^{\pi}\cos(k'/4+\theta/2)$$
$$\times\cos(-k'/4-\theta/2)A$$
$$=2^d[\cos^2(k'/4)+\sin^2(k'/4)]A\ , \qquad \text{(A6)}$$

from which Eq. (4.5a) follows.

[1]P. B. Visscher, this issue, following paper, Phys. Rev. B 29, 5472 (1984).

[2]M. Niemeijer and J. M. J. van Leeuwen, in *Phase Transitions and Critical Phenomena,* edited by C. Dohm and M. S. Green (Academic, New York, 1976), Vol. 6.

[3]J. Gunton, in *Dynamical Critical Phenomena and Related Topics,* edited by C. P. Enz (Springer, Berlin, 1979).

[4]B. I. Halperin, P. C. Hohenberg, and S.-k. Ma, Phys. Rev. B 10, 139 (1974); P. C. Hohenberg and B. I. Halperin, Rev. Mod. Phys. 49, 435 (1977).

[5]G. F. Mazenko, M. J. Nolan, and O. T. Valls, Phys. Rev. B 22, 1263 (1980).

[6]Y. Achiam and J. M. Kosterlitz, Phys. Rev. Lett. 41, 128 (1978).

[7]The notion that the convolution of a Gaussian probability distribution with itself is similar to the original distribution is actually a very old one in the theory of probability; it is what statisticians mean when they say the Gaussian distribution is "stable." See, for example, W. Feller, *An Introduction to Probability and Its Application* (Wiley, New York, 1957), p. 166. The notion is discussed in the context of critical phenomena by M. Cassandro and G. Jona-Lasinio, Adv. Phys. 27, 913 (1978).

[8]P. B. Visscher, J Stat. Phys. 25, 211 (1981).

[9]J. Kiefer and P. B. Visscher, in *Molecular-Based Study of Fluids,* edited by A. Mansoori and J. M. Haile (American Chemical Society, New York, 1983).

[10]Previous discrete hydrodynamic work (Refs. 8, 9, and 11) has used discrete transfers (analogs of fluxes) as well as contents. These are unnecessary in the present case, and may create numerical difficulties (see Ref. 9).

[11]P. B. Visscher, J. Stat. Phys. 18, 59 (1978).

[12]P. B. Visscher, Physica 108A, 152 (1981).

[13]S. Chapman, T. G. Cowling, and D. Burnett, *The Mathematical Theory of Non-Uniform Gases* (Cambridge University Press, London, 1970).

[14]J. R. Dorfman, in *Fundamental Problems in Statistical Mechanics III,* edited by E. G. D. Cohen (North-Holland, Amsterdam, 1975).