Report No. LD/78/01 (unpublished).
[5]G. McCall, private communication.
[6]A. B. Langdon and B. F. Lasinski, in Fusion Research, edited by J. Killeen, Vol. 16 of Methods in Computational Physics, edited by B. Alder, and S. Fernbach, and M. Rotenberg (Academic, New York, 1976), p. 327.
[7]D. Biskamp and H. Welter, Phys. Rev. Lett. 34, 312 (1975).
[8]C. S. Liu and M. N. Rosenbluth, Phys. Fluids 19, 967 (1976).
[9]B. F. Lasinski and A. B. Langdon, Bull. Am. Phys. Soc. 22, 1174 (1977), and to be published.
[10]C. S. Liu, M. N. Rosenbluth, and R. B. White, Phys. Fluids 17, 1211 (1974); H. H. Klein, W. M. Manheimer, and E. Ott, Phys. Rev. Lett. 31, 1187 (1973); J. F. Drake and Y. C. Lee, Phys. Rev. Lett. 31, 1197 (1973).
[11]J. M. Dawson and C. Oberman, Phys. Fluids 6, 394 (1963).
[12]H. H. Chen and C. S. Liu, Phys. Rev. Lett. 39, 881 (1977).
[13]M. D. Rosen *et al.*, Lawrence Livermore Laboratory Report No. UCRL-80862, 1978, and to be published.
[14]K. Estabrook and W. L. Kruer, Phys. Rev. Lett. 40, 42 (1978); D. W. Forslund, J. M. Kindel, and K. Lee, Phys. Rev. Lett. 39, 284 (1977).

Structure Functions of Quenched Off-Critical Binary Mixtures and Renormalizations of Mobilities

Hiroshi Furukawa
Department of Physical Sciences, Faculty of Education, Yamaguchi University, Yamaguchi 753, Japan
(Received 14 May 1979)

Structure functions of quenched off-critical binary mixtures are studied from a unified viewpoint. Individual systems are characterized only by the properties of their mobilities. The comparisons of theoretical predictions with experiments on the kinetic Ising spin model, binary fluid mixtures, and alloys yield with good agreement.

The phase separation of a binary mixture proceeds accompanied by cluster coagulations. A state where clusters are formed is represented by a local minimum of the free energy at which the first derivative of the free energy should vanish. The free energy thus contains an infinitely large number of local minima characterized by cluster sizes and cluster configurations. If the phase separation of a quenched binary mixture proceeds into a completely phase-separated state only through the local minima of the free energy, then the restoring forces acting on fluctuations with wave numbers smaller than the inverse cluster diameter should always be vanishingly small. For off-critical quenchings two further simplifications can be found. First, the fluctuations inside clusters can be neglected and therefore the restoring forces acting on fluctuations with wave numbers larger than the inverse cluster diameter are extremely large. Second, the length scale is the average cluster diameter R, only. Therefore, the structure function $S_k(t)$ may be scaled as

$$S_k(t) = R^d \tilde{S}(kR), \tag{1}$$

where d is the dimensionality. On the basis of these ideas we have derived the equation of the motion for $S_k(t)$ in the case of off-critical quenching[1] and obtained a good agreement with computer simulations on the spin-exchange kinetic Ising spin model.[2,3] Nevertheless, there remains an ambiguity in determining the R dependence of the mobility, i.e., the renormalization of the mobility is not yet considered. This is the reason why our previous discussion failed to explain other cases, e.g., the off-critical quenching of a binary fluid mixture.[4] In this short communication the renormalization of the mobility will be considered with the help of the cluster dynamics due to Binder and Stauffer.[5]

Consider the following Langevin-type equation for the composition fluctuation $\eta_k(t)$, which is the Fourier component of the local composition $\eta(t,r)$:

$$\frac{d}{dt}\eta_k = -\Gamma_k(t)\eta_k(t) + g_k(t), \quad \langle g_k(t)\eta_k{}^*\rangle_0 = 0; \tag{2}$$

$$\langle g_k(t) g_k{}^*(s)\rangle_0 = 2k_B T M(t) k^2 \delta(t-s), \tag{3}$$

where $M(t)$ is the renormalized mobility, k_B is Boltzmann's constant, T is the temperature, B is a constant, and $\langle\rangle_0$ means the ensemble average in a state observed. Then the damping coefficient $\Gamma_k(t)$ may be approximately given, for the reason mentioned above, by

$$[k_B T M(t) k^2]^{-1}\Gamma_k(t) \equiv \chi_k{}^{-1}(t) = B^{-1}R^{-d}(kR)^{d+1}. \tag{4}$$

The meaning of the exponent $(d+1)$ will be discussed below. The damping coefficient $\Gamma_k(t)$ is related to the strength of the fluctuating force, $2k_BTM(t)k^2$, through the generalized fluctuation-dissipation theorem[1,6] which can be transformed into the equation of motion for the structure function $S_k(t)$:

$$\frac{d}{dt}S_k(t) = 2k_BTM(t)k^2[1-\chi_k^{-1}(t)S_k(t)],$$

$$S_k(t) \equiv \langle|\eta_k(t)|^2\rangle_0. \quad (5)$$

Here, $\chi_k(t)$ corresponds to the structure function in a stationary state where the cluster diameter R is fixed to be constant. Thus $\chi_k(t)$ in (4) must satisfy the same scaling property as $S_k(t)$ in (1). The exponent $-(d+1)$ is so chosen that the asymptotic solution $S_k \approx \chi_k$ for large wave numbers ($kR > \pi$) may give the correct pair correlation function, i.e., $\langle\eta(t,r)\eta(t,0)\rangle_0 \propto R^{-1}r + \text{constant}$, for small r.[1]

The dependence of R on t is determined so that (1) should be the solution of (5). Thus R must satisfy the equation;

$$dR^d/dt \propto R^{-2}M(t). \quad (6)$$

So far the discussion does not rest on any particular models. If we put M equal to a constant, then (6) gives

$$R \propto k_m^{-1} \propto t^{a'}, \quad a' = (d+2)^{-1}, \quad (7)$$

where k_m is the wave number at which $S_k(t)$ has its maximum. Equation (7) agrees with the computer simulation on the spin-exchange kinetic Ising model at the late stage of coagulation for off-critical quenching.[2,3] However, there is no sure reason why we may choose a constant mobility. In fact, $a' = (d+2)^{-1}$ does not agree with that of the binary fluid mixture for off-critical quenching.[4] Although the damping coefficient $\Gamma_k(t)$ given by (4) is partially renormalized in the sense that the flatness of the free energy due to the cluster formations is already taken into account, the mobility M is not yet renormalized if we put it equal to a constant.

Let us here separate the mobility M into two parts, i.e., the bare mobility M_0 and the correction M_1 due to the cluster formation:

$$M(t) = M_0 + M_1(t). \quad (8)$$

We shall evaluate the R dependence of $M_1(t)$ on the basis of the cluster kinetics. Let D denote the cluster diffusivity which is the diffusion coefficient of the center of mass of a cluster. Then, $M_1(t)$, which is the mobility originating from the collective movement of atoms transported by a cluster of the volume R^d, can be found to be

$$k_BTM_1(t) \approx R^dD. \quad (9)$$

Equation (9) can be also found in another way; The cluster diffusivity D may be identified with $\Gamma_k^{(1)}k^{-2} \equiv k_BTM_1(t)\chi_k^{-1}$ at $k \approx \pi R^{-1}$,

$$k_BTM_1(t) \approx B\pi^{-d-1}R^dD. \quad (9')$$

This rough estimation thus gives

$$B \approx \pi^{d+1}. \quad (10)$$

Here we shall remark on the origin of M_1. Let $\eta_k(t)$ obey the nonlinear equation

$$\frac{d}{dt}\eta_k(t) = G(\{\eta\}) + f_k(t), \quad \langle G(\{\eta\})f_k^*(s)\rangle_0 = 0, \quad (11)$$

$$\langle f_k(t)f_k^*(s)\rangle_0 = 2k_BTM_0k^2\delta(t-s), \quad (12)$$

where G is a nonlinear functional of $\{\eta\}$, which is suitably chosen in each problem. Such nonlinear equations as (11) are studied for critical quenchings by transforming them into Fokker-Planck equations.[7,8] From (2), (11), and (3) we obtain

$$M_1(t) = (1/k_BTk^2)\int_0^t\langle[G(t)+\Gamma_k(t)\eta_k(t)][G(\tau)+\Gamma_k(\tau)\eta_k(\tau)]^*\rangle_0 d\tau. \quad (13)$$

We shall not, however, deal with (13) in this communication. Binder and Stauffer evaluated the cluster diffusivities of various systems.[5] Using their estimations for D, we find that $M_1 \propto R^{-1}$ for the off-critical kinetic Ising spin model and $M_1 \propto R^{d-1}$ for the off-critical binary fluid mixture. Thus, in the large-R limit we find $M_0 \gg M_1$ for the kinetic Ising model and $M_0 \ll M_1$ for the fluid. Thus there is no effect of the renormalization of M for the kinetic Ising spin model in the large-R limit, while the effect of the renormalization is dominant for the fluid. Let M be written in the large-R limit as

$$M(t) = AR(t)^{-\zeta}. \quad (14)$$

Then, $\zeta = 0$ for the off-critical Ising spin model and $\zeta = 1-d$ for the off-critical fluid. We can then find from (6) that a' in (7) is replaced by

$$a' = (d+\zeta+2)^{-1}. \quad (15)$$

For the kinetic Ising model, i.e., $\zeta = 0$, (15)

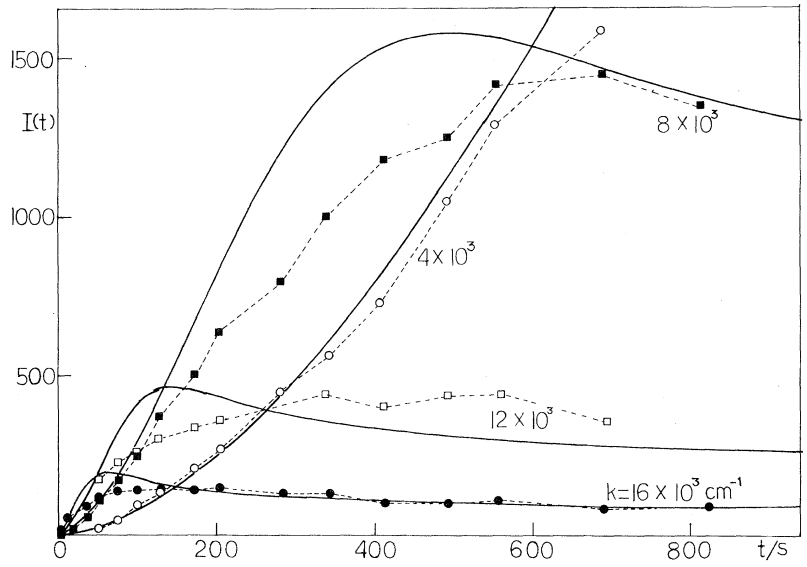

FIG. 1. $S_k(t)$ or the scattering intensity $I_k(t)$ for the off-critical quenching of isobutyric acid+water mixture. Experimental data are taken from Fig. 10 of Ref. 4. Solid curves represent $S_k(t)$ calculated by (5) for $\zeta = -2$ and therefore $a' = \frac{1}{3}$, which corresponds to the value at the late stage of coagulation. The proportionality constant of R is included in the adjustable parameter A and B.

agrees with (7). At the early stage where $M_0 \ll M_1$, however, the exponent a' is given by that of Binder and Stauffer,[5] $(d+3)^{-1}$. For the off-critical binary fluid mixture we obtain $a' = \frac{1}{3}$ in agreement both with the theoretical prediction by Binder and Stauffer[5] and with experimental observation.[4] At the early stage, however, we obtain a different value for a', i.e., $(d+2)^{-1}$.

It was previously shown[1] that $S_k(t)$ calculated by (5) for $a' = \frac{1}{5}$ in three dimensions agrees well with

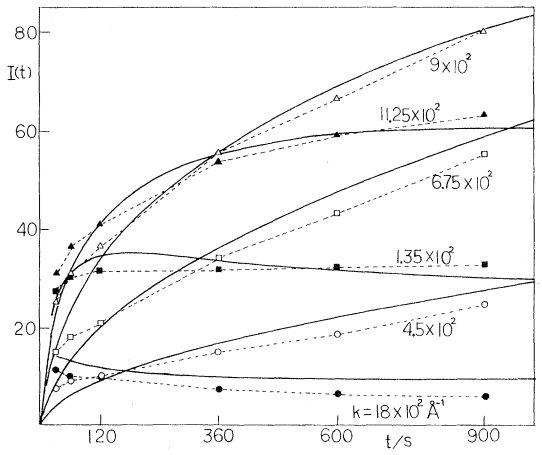

FIG. 2. $S_k(t)$ or the scattering intensity $I_k(t)$ for the off-critical quenching of Au 60% at. Pt at $T = 550°C$ ($T_c = 1270°C$). The composition lies at the center of the miscibility gap. Solid curves represent $S_k(t)$ calculated from (5) for $\zeta = 4$ and therefore $a' = \frac{1}{9}$. The adjustable parameters A and B are suitably chosen. The experimental data are taken from Fig. 3(a) of Ref. 9.

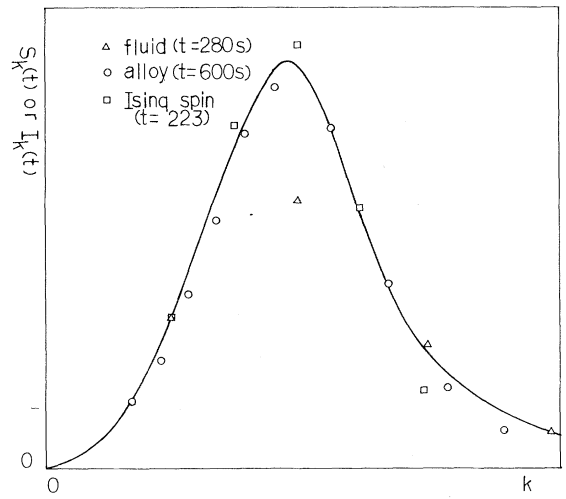

FIG. 3. Scattering intensities $I_k(t)$ for the fluid and the alloy corresponding, respectively, to Figs. 1 and 2 and $S_k(t)$ for the spin-exchange kinetic Ising model taken from Fig. 8 of Ref. 2, at various instants as functions of k. For the kinetic Ising model ζ is chosen as that of the late stage of coagulation, i.e., $\zeta = 0$, and constants A and B are suitably chosen. Arbitrary units are taken both for k and $I_k(t)$ or $S_k(t)$. The solid curve represents $S_k(t)$ calculated by (5). The theoretical profile of $S_k(t)$ does not depend explicitly on A, B, and ζ.

computer simulations on the spin-exchange kinetic Ising model[2,3] quenched at $T=0.59T_c$. We can then find that $M=M_0$ is a good approximation, since we used the unit from Langer, Bar-on, and Miller,[7] which corresponds to $M=M_0$. Furthermore, for the three-dimensional system of one-to-one composition, one may evaluate $S_k(t)\approx\chi_k(t)$ for large wave numbers ($k>k_m$) as $S_k\approx 24\pi R^{-1}k^{-4}\approx 24k_m k^{-4}$, by assuming that clusters are simple curves with R as the one side length. Here we have put $k_m\approx\pi R^{-1}$. This estimation is in good agreement with that observed in the computer simulation[2] at $T=0.59T_c$, i.e., $S_k\approx 25k_m k^{-4}$. Notice that this estimation also agrees well with $\chi_k(t)\approx\pi^3 k_m k^{-4}$ which is given with the use of (10). Thus, for the spin-exchange kinetic Ising model, even constants A in (14) and B in (4) may be determined if $k_m(t)$ is determined self-consistently. In Fig. 1, $S_k(t)$ calculated by (5) is compared with the scattering intensity from a binary fluid mixture for the off-critical quenching.[4] Two constants A and B are suitably chosen. The discrepancy between the theory and experiment at intermediate times corresponding to $k_m(t)\approx k$ is due to the over simplification of χ_k. In Fig. 2, $S_k(t)$ given by (5) is compared with the recent experiment on a quenched Au-Pt alloy.[9] The constants A and B are suitably chosen. We have put $\zeta=4$, which gives $a'=\frac{1}{9}$. However, the experimentally observed value for a' changes with time. Furthermore, the scaling property (1) seems not to be satisfied. But we consider that the experimentally observed cluster diameter R at a late stage of the coagulation would recover more rapid growth such as $R\propto t^{1/5}$, together with the scaling property (1). In Fig. 3 we compare $S_k(t)$ at various instants with experiments corresponding to Figs. 1 and 2 together with the computer simulation on the spin-exchange kinetic Ising model.[2] We note that the theoretical profile of $S_k(t)$ does not depend on ζ, A, and B. For instance, if we change the independent variable from t to R, then we obtain a ζ-independent equation for $S_k(t)$.

[1]H. Furukawa, Prog. Theor. Phys. 59, 1072 (1978), and Phys. Lett. 66A, 60 (1978).
[2]J. Marro, A. B. Bortz, M. H. Kalos, and J. L. Lebowitz, Phys. Rev. B 12, 2000 (1975).
[3]A. Sur, J. L. Lebowitz, J. Marro, and M. H. Kalos, Phys. Rev. B 15, 3014 (1977), and references quoted therein.
[4]N. C. Wong and C. M. Knobler, J. Chem. Phys. 69, 725 (1978).
[5]K. Binder and D. Stauffer, Phys. Rev. Lett. 33, 1006 (1974).
[6]H. Furukawa, Prog. Theor. Phys. 56, 464 (1976), and 58, 1127 (1977).
[7]J. S. Langer, M. Bar-on, and H. D. Miller, Phys. Rev. A 11, 1417 (1975).
[8]K. Kawasaki and T. Ohta, Prog. Theor. Phys. 59, 362 (1978).
[9]S. P. Singhal, H. Herman, and G. Kostorz, J. Appl. Cryst. 11, 572 (1978).

Normal Fluid Density of Liquid ^{3}He-B

C. N. Archie,[a] T. A. Alvesalo,[b] J. D. Reppy, and R. C. Richardson
Laboratory of Atomic and Solid State Physics and Materials Science Center, Cornell University, Ithaca, New York 14853
(Received 15 March 1979)

Measurements of the normal fluid density of ^{3}He-B from a high-Q oscillator are reported which cover the pressure range 2 to 29 bars. These results indicate that to the accuracy of the measurements, 2% in temperature and 4% in density, all pressure dependence can be accounted for by a standard weak-coupling Fermi-liquid correction. The resulting stripped normal fluid density, however, displays a significant strong-coupling contribution which conflicts with theory and the interpretation of several other experiments.

In many respects the hydrodynamics of liquid ^{3}He-B can be viewed in terms of a two-fluid model quantitatively similar to the BCS theory for s-wave superconductivity. Though quasiparticle pairing occurs in a p-wave state, the energy gap is isotropic unlike the situation for ^{3}He-A. However, complications arise even for ^{3}He-B because the pairing phenomenon itself alters the interaction responsible for the pairing. Current theoretical interest is directed towards understanding this so-called strong-coupling effect.[1-3] The superfluid density, or equivalently the den-

