# ON THE DYNAMICS OF PHASE SEGREGATION IN QUENCHED BINARY ALLOYS

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# I. INTRODUCTION

It has been made clear at this meeting that the study of kinetic phenomena in a system undergoing a phase transition has become a very popular subject during the last few years. This is not surprising. First, because both kinetic theory and phase transitions theory are challenging areas in physics so that its combination is even more particularly exciting to the theoretical physicist. Second, because also many situations arise in practice where such a combination is needed. both phenomena occur together One of the simplest situations of this kind occurs arises when a binary alloy such as Aℓ-Zn is quenched from the melt into the miscibility gap.

The theoretical analysis of this particular process, variously described as nucleation, spinodal decomposition, coarsening, and Ostwald ripening, is based mainly on the classical works of Cahn and Hilliard[1] and of Lifshitz and Slyzov[2]. The former work, as formulated by Cook[3], describes the evolution of the structure function $S(\vec{k},t)$ while the latter considers the grain distribution $n(\ell, t)$; here t is the time since quenching, $\vec{k}$ is a reciprocal wave vector, and $\ell$ is a grain size. This division corresponds directly to the two principal experimental methods of study: x-ray (or neutron) scattering for $S(\vec{k}, t)$ and electron microscopy for $n(\ell, t)$. Those classic works have been the subject of considerable study, criticism, extension, etc., in recent years; see e.g. the work of Langer[4], Binder[5], and others. These studies have made use of computer simulations of this process in simple model systems carried out by the authors and others[7]. A particularly striking feature of these computer experiments, observed by us recently[8], is the scaling behaviour of the structure function, $\tilde{S}(\vec{k},t) \sim [K(t)]^{-3} F(k/K(t))$. Such scaling was also suggested independently on theoretical grounds by various authors[5, 6, 9].

More recently, a similar scaling behavior[10] has

been found in a number of real quenched alloys and simple binary liquid mixtures; there is also some evidence that scaling may hold in other materials including glassy mixtures and $^3$He-$^4$He. The phenomenon therefore appears to be quite general, a fact which encourages further theoretical and experimental work.

The model system we used in our numerical simulation has been described before in detail[7]. At each site of a simple cubic lattice of $N = L^3$ sites (L=30 or L=50) there is assumed to be either an A atom or a B atom; the occupation variable of the $i^{th}$ site, $\eta(\vec{r}_i)$, takes on the values +1 (-1) when there is an A (B) atom at the lattice position $\vec{r}_i$. In the initial state a specified number, $\rho N$, of randomly chosen sites are occupied by A atoms and the rest by B atoms. This corresponds to an infinite temperature state. The relative concentration of A atoms is $\rho = (\bar{\eta} + 1)/2$, $o \leq \rho \leq 1$, where $\bar{\eta} = N^{-1} \sum_i \eta(\vec{r}_i, t)$ is the average "magnetization" in the lattice (fraction of A atoms less the fraction of B atoms), which is constant in time. The evolution proceeds by choosing, at a rate $\alpha/3$, a pair of nearest neighbor sites, i, j, in the lattice. If the sites are occupied by different kinds of atoms there is a probability (which depends on the change in the energy of the system caused by the interchange: the so-called Kawasaki dynamics[11]) that they will be interchanged. For our model system the energy is assumed to be given by the usual nearest-neighbor Ising hamiltonian. Periodic boundary conditions are assumed.

We have analysed with some details seven points $P_j$, $j = 1, \ldots, 7$, in the phase diagram of the model. $P_1$-$P_5$ correspond to $T \simeq 0.59\ T_c$ and fractional concentration of A-atoms $\rho = 0.05$, 0.075, 0.10, 0.20 and 0.50 respectively. At this temperature the value of $\rho$ at the coexistence line is $\rho_v \simeq 0.015$, $m_o \simeq 0.97$. The points $P_6$ and $P_7$ are at $\rho = 0.5$ and $T \simeq 0.78T_c$ and $T \simeq 0.89$ respectively. At $0.78T_c$, $\rho_v \simeq 0.0613$ $m_o \simeq 0.877$, while at $0.89T_c$, $\rho_v \simeq 0.1246$, $m_o \simeq 0.75$.

The quantity of primary interest in our observation was $S(\vec{k},t)$, the structure function at time t following quenching. $S(\vec{k}, t)$ is the Fourier transform of the spatial correlation function $G(\vec{r}, t)$:

$$S(\vec{k}, t) = \sum_{\vec{r}} \exp(i\vec{k} \cdot \vec{r})\, G(\vec{r}, t) \quad , \tag{1}$$

$$G(\vec{r}, t) = N^{-1} \sum_{i} \langle [\eta(\vec{r}_i, t) - \bar{\eta}\,] [\, \eta(\vec{r}_i + \vec{r}, t) - \bar{\eta}\,] \rangle \tag{2}$$

where $\vec{r}$ and $\vec{r}_i$ run over the N lattice sites and $\vec{k} = (2\Pi/L)\vec{\mu}$, $\mu_\alpha = 0, \pm 1, \ldots, \pm L/2$, $(\alpha = 1,2,3)$, specifies the first Brillouin zone. The $\langle \ldots \rangle$ represents an ensemble average which could in principle be implemented on the computer by making "many" independent runs. Note that $S(\vec{k}, t)$ is periodic in each component with period $2\Pi/L$, $S(\vec{k} = 0, t) = 0$ and

$$N^{-1} \sum_{\vec{k}} S(\vec{k}, t) = 1 - \bar{\eta}^{\,2} \quad . \tag{3}$$

In order to compare with experiments on polycrystalline materials we define an average structure function depending only on the wave number $k = 2\Pi\mu/L$, $\mu = 0,1,\ldots, \sqrt{3}\ L/2$,

$$\bar{S}(\vec{k},t) = \sum_{[\,\vec{k}\,]} S(\vec{k}, t)/(1-\bar{\eta}^2) \Big/ \sum_{[\,\vec{k}\,]} 1 \tag{4}$$

where $\sum_{[\,\vec{k}\,]}$ is the sum over all values of $\vec{k}$ in the first octant, $k_\alpha \geq 0$, $\alpha = 1,2,3$, such that

$$(2\Pi/L)\ (\mu - 1/2) \leq |\vec{k}| \leq (2\Pi/L)\ (\mu + 1/2) \quad .$$

## II. BEHAVIOR OF $\bar{S}(k, t)$

Figure 1 shows plots of $\bar{S}(\vec{k}, t)$ vs. k at different times for quenches to the point $P_1$. These plots are qualitatively similar to those obtained at other points $P_j$. At t=0, the system is completely disordered and $\bar{S}(\vec{k},0) \sim 1$ independent of k. The system is then quenched to some low temperature inside the coexistence curve. Thermal processes

Fig. 1. Development with time of $\bar{S}(\vec{k}, t)$ versus k at different times in the case of a quench of the model system with N = 125,000 sites to the phase point $P_1$. Note that, in this case, we only have $\bar{S}(\vec{k}, t)$ at forteen different values of k, $k=2\pi\mu/50$, $\mu=1-14$, which have been connected by straight lines. Increasing values of the time correspond to the different graphs from the bottom of the picture to the top. The graphs at different values of the time tend to form a common envelope for $k>k_{max}$, the location of the maximum intensity (which is shifting with time towards smaller values of k). This is in contrast with the cross-overs characterizing the tail $k>k_{max}$ of the $\bar{S}(\vec{k}, t)$ vs. k curves in the case of quenches to $P_4$ (Fig. 2, Ref.

7c) and to $P_5$ (Fig. 2, Ref. 7a).

lead to the migration of A and B atoms, proceeding (in our model) via nearest neighbor exhanges given by the transition probability mentioned above, which now drive the system towards equilibrium corresponding to segregation into A-rich and B-rich regions. As these regions grow in size, $\bar{S}(\vec{k}, t)$ develops a peak at $k = k_{max}(t) = \pi/R(t)$ where $R(t)$ represents some characteristic length in the system at time $t$ following the quench. This length will grow in time as the single phase regions grow to macroscopic sizes so that $k_{max}(t) \to 0$ as $t \to \infty$ in a macroscopic system. (In the computer simulations we obviously have $R(t) < \frac{1}{2} L$.) For contrast we also show, in Fig. 2, the qualitatively very different behavior of $S(\vec{k}, t)$ when the system is quenched to the point $P^*$, $\rho = 0.0613$, $T \simeq 0.78 T_c$, at the coexistence curve.

Fig. 2. Same as Fig. 1 in the case of a quench to $P^*$,

$T \simeq 0.78T_c$, $\rho = \rho_v(T) = 0.0613$, i.e. on the coexistence line at the same temperature as $P_6$. A comparison with Fig. 1 shows up how the model system differentiates $P^*$ from $P_1$, say. (Note that the ordinate scale differs by two orders of magnitude). Here increasing values of the time correspond to increasing values of the number labeling the different graphs.

Comparisons of the graphs for points $P_1$ to $P_5$ shows that the speed of the segregation increases, in units of attempted exchanges, with the distance of the quench from the coexistence line. The same is true for the points $P_5$ to $P_7$. This accounts, in the main, for the widely different time lengths to which we ran our simulations. It is clear that close enough to the coexistence line at $T \simeq 0'6T_c$ the relaxation time would become so long that the system would appear, for all practical purposes, to be in a metastable state while close to $T_c$ the system will be in the region of critical slowing down. In either case the segregation would not be visible in our simulations.

It should be noted that the relaxation time increases rapidly as one approaches the coexistence line. There is no evidence, however, in our simulations of any *abrupt* change in the behavior on crossing any of the proposed theoretical spinodal lines. There are some differences however in the way in which $S(\vec{k}, t)$ evolves with time in different parts of the coexistence region: for quenches deep in the coexistence region $S(\vec{k}, t)$ decreases strongly after reaching its maximum value while near the coexistence line it remains approximately constant for the times observed. This behavior is also true for other values of $k > k_{max}$ and results in "cross-overs" at large k seen in $S(\vec{k}, t)$ vs. k curves, for $P_4$ in Fig. 2a of Ref. 7c and for $P_5$ in Fig. 2 of Ref. 7a but not in Fig. 1 here. Such cross-overs have been conjectured[4] to be the hallmark of quenches inside the spinodal curve and our simulations give some evidence of

this. The evidence is however not entirely conclusive since the differences may be due to not waiting "long enough" near the coexistence line and the scatter is relatively large (see e.g. Figs. 3-4 in Ref. 7a corresponding to the quenches to $P_6$ - $P_7$). Let $\hat{S}(\vec{k}, t)$ be the limit of $S(\vec{k}, t)$ as $N \to \infty$. It is this smooth function about which we would like to obtain information from our computer simulations. Hence it is essential that we look for quantitative features in our computed $S(\vec{k}, t)$ which will go over smoothly to the macroscopic $\hat{S}(k, t)$. In order to understand the long time behavior of $\hat{S}(\vec{k}, t)$ we note that as $t \to \infty$ we expect that $\hat{S}(\vec{k}, t) \to \hat{S}_{eq}(k)$, the equilibrium structure function of a macroscopic system fully segregated into two pure phases. This is given by

$$\hat{S}_{eq}(k) = (m_o^2 - \bar{\eta}^2)\, \bar{\delta}(k) + \hat{S}_c(k; T) \qquad (5)$$

where $\delta(k)$ is the sphericalized Dirac delta function at $k = 0$, $\hat{S}_c(k; T)$ is the equilibrium structure function on the coexistence line (by the symmetry of our model system this is the same for both pure phases) and $m_o(T)$ is the spontanuous magnetization (equal to $\bar{\eta}$ on the coexistence line). As already noted, the system, after quenching, will segregate locally into regions (often referred to as grains, clusters or droplets) of A-rich and B-rich phases and then will evolve further by the growth of these segregated regions (coarsening or Ostwald ripening). We might expect that after some initial time the structure function "within" the segregated regions will be close to its equilibrium value $\hat{S}_c(k; T)$. It seems therefore reasonable to consider the quantity

$$\hat{S}_1(k, t) = \left[\hat{S}(k, t) - \hat{S}_c(k; T)\right] \quad (m_o^2 - \bar{\eta}^2)^{-1}, \qquad (6)$$

which approaches $\bar{\delta}(k)$ with time, as most relevant for the description of the coarsening process.

We shall therefore consider in our analysis also the analogous quantity for our finite system

$$S_1(\vec{k},\ t) = [S(\vec{k},\ t) - S_c(\vec{k};\ T)]\ \ (m_o^2 - \bar{\eta}^2)^{-1}, \quad (7)$$

in the hope that it will more clearly reveal the essential features of the coarsening process. The function $S_c(\vec{k};\ T)$ was obtained, for the three different temperatures T considered here, by quenching to points on the coexistence like and waiting for the system to reach equilibrium.

A quantitative feature of $S(\vec{k},\ t)$ we looked at in our simulations were the moments

$$k_n(t) = \langle k^n \rangle = \sum_{k=0}^{K} k^n S_1(\vec{k},\ t) \Big/ \sum_{k=0}^{K} S_1(\vec{k},\ t)\ ,\ n = 1,2\ , \quad (8)$$

where $K \simeq 0.55\Pi$. We found that $\langle k^2 \rangle / \langle k \rangle^2$ was essentially independent of t. This suggest that our $S_1(\vec{k},\ t)$ with discrete argument k having spacings $2\Pi/L$ might be related for late times to the macroscopic structure function $\hat{S}_1(\vec{k},\ t)$ via a smooth scaled function F such that

$$S_1(\vec{k},\ t) \simeq \hat{S}_1(\vec{k},\ t) = b(t)F(\vec{k}/\vec{K}(t)\ ) \quad (9)$$

where K(t) is some characteristic wave vector in the system and b(t) is a normalizing factor. Now if this is indeed the case, we have from Eqs. (3) and (6) that

$$\int_B k^2 \hat{S}_1(\vec{k},t)\,dk = b(t)K^3\ (t) \int x^2 F(x)\,dx = 2\Pi^2\ , \quad (10)$$

independent of t. The integration on the left side of Eq.(10) is over the first Brillouin zone and so the x integration is over a cube centered at the origin with sides of length $2\Pi/K9t)$. For large t, $K(t) \to 0$, and the integral in Eq. (10) can be taken over all space. Since F(x) can be expected to decay quite fast for large x very little error will be made even at quite early times if we set $b(t) = 2\Pi^2 K^{-3}$ and normalize F so that

$$\int_0^\infty x^2 F(x)\,dx = 1 \quad . \tag{11}$$

To test this scaling hypothesis and find the smooth $F(x)$ from our simulations we *define* a function of two variables, $x = k/k_1(t)$ and $t$, by the relation

$$F(x;t) = \frac{L}{\Pi} k_1^3(t)\, S_1(xk_1(t),t) \Big/ \sum_{k=0}^{K} k^2 S_1(k,t) \quad , \tag{12}$$

and then see whether for late times $F(x;t) \simeq F(x)$ is a smooth function of $x$ independent of $t$.

The values of $F(x;t) \simeq F(x)$ independent of $t$, obtained from our simulations for large times are shown in Figs. 3-5 for "deep", "intermediate" and "shallow" quenches. While these curves are all similar, the "experimental" scatter within each group is significantly smaller than between them. We believe therefore that there is a real, albeit small, difference between the scaling function $F(x)$ at different quenches. The similarity $F(x)$ near $T_c$ and near the coexistence curve for small $\rho$ would appear to suggest some sort of "spinodal line" criticality. This is an intriguing question which is however very difficult to answer at present. Figs. 3-5 also show the function $F(x;t)$ at some earlier times as indicated. We have also included in those figures a fit of the form $(\alpha_1 + \alpha_2 x^4)^{-1}$ to the "tail", $x > x_{max}$, of the scaling function $F(x)$. The results of our analysis of $k_1(t)$ are summarized in Table I where it is shown that the data consistent with the Lifshitz-Slyzov predictions[2, 7, 9].

## III. COMPARISON WITH EXPERIMENT

The model used in our computations certainly involves a great over-simplification of the behavior of real alloys where the processes we are interested in are greatly influenced by elastic distortions, grain boundaries, vacancies and other competing phenomena. The behavior of the model can thus only be compared with that of real materials in highly idealized conditions. Despite this, previous

TABLE I

| phase point | max. duration of run | $k_1 \sim t^{-a}$ | $k_1^{-3} = A + Bt/10^3$, $t \geq t_o$ | | |
|---|---|---|---|---|---|
| | | a | A | B | $t_o$ |
| $P_1$ $0.59T_c$ 5% | 14000 | 0.35 | 8.8 | 1.5 | 6800 |
| $P_2$ $0.59T_c$ 7.5% | 10200 | 0.23 | 7.5 | 1.5 | 4000 |
| $P_3$ $0.59T_c$ 10% | 73000 | 0.21 | 3.6 | 1.5 | 2500 |
| $P_4$ $0.59T_c$ 20% | 3900 | 0.19 | 2.3 | 1.7 | 1500 |
| $P_5$ $0.59T_c$ 50% | 650 | 0.19 | 1.2 | 3.0 | 350 |
| $P_6$ $0.78T_c$ 50% | 1700 | 0.23 | 1.4 | 3.9 | 1000 |
| $P_7$ $0.89T_c$ 50% | 6600 | 0.25 | 3.5 | 3.9 | 1000 |

Table I. Values of the adjustable parameters for two different fits to $k_1$: first assuming a simple power law (here all the data except the very early one is included in the fit), and then assuming a lineal behavior of $k_1^{-3}$ with time (using only data for $t \geq t_o$). Here $t_o$ is the approximate time at which we observed the onset of the dynamical scaling of the structure function according to Eq. (9).

Fig. 3. The scaling function $F(x)$ in the case of "deep" quenches: the triangles correspond to a quench to $P_4$ and they include times $t \geq 1500$, the asterisks to $P_5$ and $t \geq 350$, and the circles to $P_6$ and $t \geq 1000$ (see also caption for Fig. 4).
The dashed line represents the shape of the function F at an earlier time when scaling does not yet hold: it was drawn connecting the discrete values of $F(x;t)$ for $t=110$ in the case $P_4$. The dotted line corresponds to a function $F^{-1}=\alpha_1+\alpha_2 x^4$ trying to fit the experimental data for

$x > x_{max}$, the location of the maximum of F(x).

Fig. 4. The function F(x;t) defined in Eq. (12) is plotted here versus $x = k/k_1(t)$ in the case of the ("intermediate") quench to $P_3$. Every symbol (o, 1, 2,3,...) in the graph corresponds to a different value of the time: the first one (zero) is for $t \simeq 2500$, the time increasing up to $t \simeq 7300$ which is represented by crosses. All the values of F(x;t) in that time interval seem to lie on a common curve at different values of x. The dashed line was drawn connecting the values of F(x,t) at t=228. The dotted line corresponds to a fit $F^{-1} = \alpha_1 + \alpha_2 x^4$ to the "tail" $x > x_{max}$ of F(x).

Fig. 5. Same as Fig. 7 in the case of ("shallow") quenches to $P_1$ (triangles; all the data $t \geq 6800$ is included in this case), $P_2$ (asterisks, $t \geq 4000$) and $P_4$ (circles, $t \geq 1000$). The dashed line is for $P_1$ and $t_0 = 209$.

analysis have shown that our results are frequently very similar to experimental observations[7d]. In order to make comparison with experiments, however, care must be paid to the relation between the units and other characteristics in our model and those corresponding to real alloys[8].

We have analysed the data reported by Singhal, Herman and Kostorz[12] corresponding to the Au-60 at % Pt alloy quenched into iced brine; the aging at around 823°K was observed by neutron scattering during 900 sec. The composition of the sample lies at the center of the miscibility gap, and $T \simeq 0.6 T_c$ so that the experiment may be

compared with our simulations at $P_5$ and perhaps at $P_4$. The scattering analysis gives a structure function with a shape quite similar to the one observed in our simulations, including the characteristic crossovers at that temperature and composition in the tail, $k>k_{max}$. We also find that the data in Ref. 12 satisfy the scaling hypothesis in the time range 120 sec $\leq$ t $\leq$ 900 sec for all but very large x, say x>1.8, see Fig. 6. Indeed the experimental function F(x) is quite similar to the corresponding one in the case of our simulations at $P_4$ and $P_5$. In fact one can make the data from the actual and computer experiments lie on the same curve by only re-scaling the vertical axis, as seen in Fig. 6.

Fig. 6. We compare in this figure data from the computer simulation (empty symbols) with the experimental data in Fig. 3 from Ref. 12 (full symbols) corresponding to Au-60 at % Pt alloy quenched to T 0.6 $T_c$. The full circles are for t = 900 sec and the stars for t = 360 sec, both lying on the same curve except perhaps for x $\geq$ 1.8. The broken line 1 is for t = 0 (the initial sample was

already decomposed to some extent), and the broken line 2 is for t=30 sec, in both cases scaling does not yet hold. The empty circles correspond to our simulation at $P_5$ while the empty squares are for $P_4$ (see Fig. 3). Only the vertical scale in Fig. 3 needed to be changed in order to obtain the present fit.

We have found a similar agreement between our scaling function F(x) at $P_1$ and $P_2$, and the one reported by Guyot et al[13] corresponding to a sample of Aℓ-15 at % Zn quenched to $T \simeq 0.6T_c$; this is shown in Fig. 7.

Fig. 7. The data in Fig. 5 is compared with experimental data (Ref. 13) on an Aℓ-15% Zn alloy quenched to 90°C (solid line) and to 110°C (dashed line) (both T $0.6T_c$). The vertical and horizontal scales in Fig. 5 where changed to obtain the present fit; a fairly good fit can also be obtained, however, by changing only the vertical scale in Fig. 5.

Finally, recent observations on binary fluid mixtures report a behavior which is also qualitatively similar to the one shown by our model system. In particular, Goldburg's and Knobler's groups find[8] a scaling behavior of $\hat{S}(k, t)$ with a function $F(x)$ which looks quite similar to our function.

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