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Growth of Clusters in a First-Order Phase Transition

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The results of computer simulations of phase separation kinetics in a binary alloy quenched from a high temperature are analyzed in detail, using the ideas of Lifshitz and Slyozov. The alloy was modeled by a three-dimensional Ising model with Kawasaki dynamics. The temperature after quenching was $0.59T_c$, where T_c is the critical temperature, and the concentration of minority atoms was $\rho = 0.075$, which is about five times their largest possible single-phase equilibrium concentration at that temperature. The time interval covered by our analysis goes from about 1000 to 6000 attempted interchanges per site. The size distribution of small clusters of minority atoms is fitted approximately by $c_1 \approx (1-\rho)^3 w(t)$, $c_l \approx (1-\rho)^4 Q_l w(t)^l$ $(2 \le l \le 10)$; where c_l is the concentration of clusters of size l; $Q_2, \dots, Q_{10}$ are known constants, the "cluster partition functions"; t is the time; and $w(t) = 0.015(1 + 7.17t^{-1/3})$. The distribution of large clusters $(l \ge 20)$ is fitted approximately by the type of distribution proposed by Lifshitz and Slyozov, $c_l(t) = -(d/dl)\psi[\ln t + \varphi(l/t)]$, where φ is a function given by those authors and ψ is defined by $\psi(x) = C_0 e^{-x} - C_1 e^{-4x/3} - C_2 e^{-5x/3}$; C_0, C_1, C_2 are constants determined by considering how the total number of particles in large clusters changes with time.

KEY WORDS: Nucleation; cluster growth; phase separation.

1. INTRODUCTION

The basic features of the phase segregation process in a binary alloy such as Zn–Al, following quenching into the miscibility gap, can be simulated on a

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computer for a simple model of such a system. In this model, which has been described extensively elsewhere,(1–9),[6] each site of a (simple cubic) lattice is occupied by either an A or B particle. Particles on nearest neighbor sites interact in a way which favors segregation. As is well known, this alloy model is isomorphic to a lattice gas with nearest neighbor attractive forces in which the phase segregation is into a liquid and a vapor phase. (This is accomplished by identifying sites occupied by B particles with empty sites and A particles with particles of the lattice gas.)

The microscopic kinetics of this system is represented(1) by Kawasaki dynamics, a Markov process whose basic step, in lattice-gas language, is to move one particle to a neighboring empty site. More specifically, at each step of the simulation a site is chosen at random, then a neighboring site is chosen at random, and then if the two sites are occupied by different kinds of particle (or in lattice-gas language, if one is occupied and the other empty), their occupation numbers are interchanged with a probability, chosen to satisfy detailed balancing,

$$p(\Delta E) = e^{-\Delta E/2kT} F(\Delta E) \tag{1}$$

Here ΔE is the increase of energy brought about by the proposed interchange and F is an even function, which in this work was taken to be $[2\cosh(\Delta E/2kT)]^{-1}$. The number of times that this process is attempted, divided by the total number of sites, is taken as our measure of the time.

The phase diagram of this system is accurately known from series expansions. The critical temperature T_c is very close to $1.13V/k$, where V is the amount of energy required to separate two A particles on adjacent lattice sites and k is Boltzmann's constant.(10)

In a recent paper(9) we discussed a series of simulations made at $T = 0.59T_c$ [i.e., $(V/kT) = 1.5$ exactly] for various small values of ρ, the fraction of sites occupied by A particles (corresponding, in the lattice-gas interpretation, to the fraction of occupied sites). The smallest value of ρ was 0.0146, the value in the saturated B-rich phase (corresponding to saturated vapor). The other values used for ρ were 0.02, 0.035, 0.05, 0.06, 0.075, and 0.1. The initial state was chosen to be random, corresponding to quenching from an infinite temperature.[7] The lattice was simple cubic with $50 \times 50 \times 50$ sites (or in a few cases $30 \times 30 \times 30$ sites) and periodic boundary conditions.

The analysis in Ref. 9 dealt with equilibrium and quasiequilibrium properties, particularly the distribution of cluster sizes, and revealed the probable existence of metastable states at densities ρ below about 0.035. In

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[6] For early simulation work of this type see Ref. 2.

[7] In a few cases the system was first brought to equilibrium at the saturation temperature for the density ρ and then quenched to the lower temperature. Except for the behavior at very early times, the results appeared to be the same.

the present paper we analyze a single, very long run at density $\rho = 0.075$, but the qualitative behavior is similar for densities 0.05, 0.06, and 0.1. The behavior of the structure function, energy, and some gross cluster properties at high densities, $\rho = 0.2$ and 0.5, is described in Refs. 4–8, and at the lower densities in Ref. 11.

2. QUALITATIVE BEHAVIOR

The time evolution of the system following quenching can be broken up into three stages. In the first stage, which lasts for about ten time units (that is, ten attempted interchanges per site) the distribution of clusters changes rapidly, away from its initial form in which nearly all the particles are in monomers (one-particle clusters), toward one with fewer monomers and many more large clusters of sizes from two upward.

During the second stage, which lasts from about the 10th to the 100th time unit, the number of large clusters (larger than about 30 particles) continues to grow, but the distribution of small clusters has stabilized to some extent; in fact this distribution can be fitted quite well, for $l \leqslant 10$, by the empirical formula obtained in Ref. 9:

$$c_1 = (1 - \rho)^3 w, \qquad c_l = (1 - \rho)^4 Q_l w^l \quad (2 \leqslant l) \tag{2}$$

where c_l ($l = 1, 2,\ldots$) denotes the number of l-particle clusters per lattice site, w is an adjustable parameter, whose value changes with time as the cluster distribution changes, and Q_l is the partition function for l-particle clusters, defined in Ref. 12 as

$$Q_l = \sum_K{}' e^{-\beta E(K)} \tag{3}$$

In this formula the sum contains one representative from each translational equivalence class of l-particle clusters, and E denotes the energy of the cluster K, which is equal to $-V$ times the number of adjacent pairs of occupied sites in it.

By about 125 time units the number of large clusters has ceased to grow. This signals the beginning of the third stage, in which the number of large clusters no longer grows, but decreases slowly. Most of the clusters that remain are growing in size, however, and as a result the total number of particles in large clusters increases slowly despite the decreasing number of such clusters. At the same time the value of w in formula (2) falls slowly because the number of particles in small clusters is decreasing. Even by the end of our longest run, however, which was at 6467 time units, the value of w and the number of particles in small clusters are well above the values they would be expected to take at equilibrium, and there are still about 20 large clusters present with sizes ranging from 25 to 750, in place of the single large cluster of size about 7500 that one would expect to find at equilibrium.

Table I compares the observed cluster concentrations c_l with those calculated from the empirical formula (2). In each pair of rows the observed values are given in the upper row and the calculated values in the lower row. Each value given for the time t is the average of 20 successive times at which "observations" of the cluster distribution (i.e., the values of the c_l) were made and recorded by the computer. From each set of 20 cluster distributions an average distribution $c_1, c_2,\ldots$ was calculated and is recorded in the upper row of a pair. The values of $c_1, c_2,\ldots$ shown in the lower row were calculated from the empirical formula (2) with w chosen to make the observed and calculated values of $\sum_{l=1}^{10} lc_l$ equal. A comparison of the upper and lower row in each pair shows that the values of c_l for $1 \leqslant l \leqslant 10$ are given quite well by formula (2) with w depending on the time. The finding that these values of c_l depend on time only through w confirms the ideas discussed qualitatively in Ref. 9 and is in agreement with a suggestion by Binder: see Eq. (3.8) of Ref. 13.

A mathematical description of the behavior we observed has been given by Lifshitz and Slyozov.(14) The physical basis of their theory rests on the fact that the vapor pressure over a curved surface exceeds that over a plane surface by an amount proportional to $1/R$, where R is the radius of curvature. Extrapolating this idea to values of R as small as the radius of a cluster and taking the clusters to be roughly spherical, one assumes that the "vapor pressure" of a cluster of size l exceeds the saturated vapor pressure by an amount proportional to $l^{-1/3}$, and hence that near a cluster of size l the steady-state value of w exceeds its value w_s for the saturated vapor by an amount proportional to $l^{-1/3}$. If the actual value of w exceeds the vapor pressure of some cluster, that cluster is likely to grow; but if w is less than the vapor pressure of the cluster, it is likely to shrink.

At the beginning of the second stage of the process described above, the value of w is larger than the vapor pressure of most of the large clusters (which have been formed by fluctuations) and so most of them grow. This increases the number of particles in large clusters and so reduces the number in small clusters and hence reduces the value of w. Eventually w becomes less than the vapor pressure of some of the smaller of the "large" clusters. These will then tend to shrink instead of growing further. The lowered value of w also makes it difficult for any new large clusters to form by fluctuations. In fact, from this time on, the number of large clusters will tend to decrease: the third stage of the process has begun.

During this third stage w tends to adjust itself to a value such that the growth of large clusters just balances the shrinking of small clusters; for, if w is too large, then most of the large clusters will grow, so that the number of

Table I. Observed Cluster Concentrations c_l Compared with the Empirical Formula (2)

t	w	$10^5 c_1$	$10^5 c_2$	$10^5 c_3$	$10^5 c_4$	$10^5 c_5$	$10^5 c_6$	$10^5 c_7$	$10^5 c_8$	$10^5 c_9$	$10^5 c_{10}$
1189		1399	306	116	61	33	22	15	10	7	6
	0.01758	1391	304	120	61	35	22	14	10	7	5
2121		1337	278	101	49	27	16	10	6	5	4
	0.01675	1326	276	104	50	27	16	10	7	5	3
3137		1277	253	88	42	18	11	7	5	3	2
	0.01593	1261	250	89	41	21	12	7	5	3	2
4250		1215	233	72	32	14	9	4	3	2	1
	0.01512	1196	224	76	33	16	9	5	3	2	1
5431		1210	224	66	28	13	6	4	2	1	1
	0.01480	1172	216	72	31	15	8	4	3	2	1

particles in large clusters increases, the number in small clusters decreases, and so w decreases; and if w is too small, then most of the large clusters will shrink, transferring particles from large to small clusters and increasing w. Thus the only changes in w are henceforth comparatively slow ones, brought about by slow changes in the distribution of large clusters. One of the most important processes changing this distribution is the steady disappearance of the smallest "large clusters" from it as they eventually become too small to qualify as large clusters any more. This process tends to increase the average size of the remaining large clusters, and so to reduce the value to which w adjusts itself. In consequence the value of w decreases slowly, and so does the total number of particles in small clusters.

The basic processes in this mechanism are the condensation of a small cluster, typically a monomer, onto a large cluster, and its inverse, the evaporation of a small cluster from a large cluster. The rate at which these processes change the sizes of the large clusters depends on the diffusion of small clusters.(14) Other processes, however, can also change the size of a large cluster, and should in principle be included in the discussion. Typical of such processes are the coagulation of two large clusters to form an even larger one, and its inverse, the breaking up of a large cluster into two large parts. Rough estimates indicate that processes involving more than one large cluster are relatively unimportant at sufficiently large times.

One reason for this, stated simply, is that D_l, the diffusion constant of a large cluster of size l, will decrease for large l, e.g., $D_l \sim l^{-\gamma}$ ($\gamma \sim 1$ in two dimensions,(6) $\gamma \sim 2/3$ in three dimensions(15)). The relative effects of small and large clusters on the transport of matter by diffusion will be in proportion to the diffusion constant multiplied by the number of particles in such clusters. As time proceeds, the number of particles in small clusters decreases only very slowly, and the number in large clusters increases correspondingly; their ratio multiplied by the number $l^{-\gamma}$ measuring the ratio of their diffusion constants gets smaller and smaller, and so for large enough times we may expect to be able to ignore transport by diffusion of large clusters.

Even if their centers of gravity do not move, two large clusters can coagulate if their growth brings their surfaces into contact. If c large clusters per site start growing at random positions, a cluster that reaches the size l will, on the average, have absorbed lc other large clusters on the way (all the ones whose points of origin are now within it). Even the largest of these absorbed clusters cannot have been larger than $\frac{1}{2}l$ at the moment of absorption, and their average size at the moment of absorption may be estimated as half of this, which is $\frac{1}{4}l$. Therefore the number of particles in the l-particle cluster that were obtained by absorption of other large clusters is roughly $\frac{1}{4}l \cdot cl$. The ratio of this to the current size of the cluster is about $\frac{1}{4}cl$, which is $\frac{1}{4}$ of the number of particles per site in large clusters and is therefore less than

2% in the run made at $7\frac{1}{2}\%$ density. As a first approximation, therefore, we may neglect the growing together of large clusters, as well as their diffusion motion, in comparison with the mechanism based on interactions between large clusters and small ones.

Although we have not taken interactions between large clusters into account in our analysis, these interactions did have a noticeable effect, particularly on the very large clusters. For example (see Table VIII in Appendix B) at time 1981 the largest cluster contained 285 particles, but on the next observation, at time 1997, the largest cluster contained 441 particles, having apparently been formed through the coagulation of two clusters each containing about 220 particles. Effects of this kind are discussed by Lifshitz and Slyozov(14) as a correction to their theory, and by Binder and co-workers as an integral part of the kinetic description.(16–18) They may be expected to increase in importance as the density increases,(1–8) because at higher densities there are more particles in large clusters.

3. QUANTITATIVE BEHAVIOR: KINETIC EQUATIONS

A number of different systems of kinetic equations have been used for the discussion of the type of problem we are considering. The best established are the equations of Becker and Döring, in which the only processes considered are the absorption and the emission of a monomer by a cluster of arbitrary size. The equations for c_l, the mean density of clusters of size l, are in Refs. 19 and 20,

$$dc_l/dt = J_{l-1} - J_l \qquad (l \geqslant 2) \tag{4}$$

where J_l, the net rate of conversion (per unit volume or per site) of l-particle to $(l + 1)$-particle clusters, is given by

$$J_l = a_l c_l c_1 - b_{l+1} c_{l+1} \qquad (l \geqslant 1) \tag{5}$$

Here a_l is a coefficient describing the rate at which l-particle clusters absorb monomers and b_{l+1} is a coefficient describing the rate at which $(l + 1)$-particle clusters emit monomers. To complete the system of equations we also need one for c_1. The condition determining c_1 is the conservation of matter, which can be written

$$\sum_{l=1}^{\infty} l c_l = \rho = \text{const} \tag{6}$$

The coefficients in the formula (5) for J_l are related through the fact that, by detailed balancing arguments, $J_l = 0$ at equilibrium. If the density is small enough for the equilibrium state to have only one phase, it is reasonable to assume that the equilibrium cluster concentrations c_l are given approximately by (2) for all l. Substituting from (2) into (5) and setting $J_l = 0$, we obtain for the ratio b_{l+1}/a_l a formula which depends on the density only weakly, through the factor $(1 - \rho)^3$:

$$b_{l+1}/a_l = [c_l c_1/c_{l+1}]_{\mathrm{eq}} = w_l(1 - \rho)^3 \qquad (l \geqslant 2) \tag{7}$$

where

$$w_l = Q_l/Q_{l+1} \tag{8}$$

The factor $(1 - \rho)^3$ represents the reduction in the probability of evaporation of a monomer from an $(l + 1)$-particle cluster caused by the possibility that a monomer cannot (by definition) form on any site next to a site that is already occupied. We shall assume that (7) is valid also at higher densities, for which the equilibrium state has two phases, although (2) is clearly not valid here for large l either in the true equilibrium state or even in the quasistationary state with a time-dependent w.

For $l \leqslant 9$, the exact values of w_l are available.(12,21) They are shown in Table II for $T = 0.59T_c$. The last row of the table gives values of the empirical formula

$$w_l \approx w_s[1 + 2.415/(l - 2)^{1/3}] \qquad (l \geqslant 3) \tag{9}$$

where

$$w_s = 0.010526 = \lim_{l \to \infty} w_l \tag{10}$$

is the value of w for the saturated vapor, whose value we know from previous work.(9) The physical basis for the approximate formula (9) is the idea mentioned earlier that the "vapor pressure" over a droplet of size l exceeds that over a plane surface by an amount proportional to the curvature of the surface of the droplet. Equation (9) provides a convenient way of extrapolat-

Table II. Exact Values of w_l Compared with the Approximation (9) at $T = 0.059T_c$

l	1	2	3	4	5	6	7	8	9
w_l	0.0744	0.0446	0.0347	0.0307	0.0282	0.0266	0.0254	0.0245	0.0238
Approximation	—	—	0.0359	0.0307	0.0282	0.0265	0.0254	0.0245	0.0238

ing w_l to values of l greater than 9. In the earlier stages of this work, a different approximation, of the form

$$w_l \approx w_s \exp[\mathrm{const}/(l - \mathrm{const})^{1/3}]$$

suggested by one of the extrapolation formulas for Q_l discussed in Ref. 9, was used, but this approximation formula is less accurate for $3 \leqslant l \leqslant 9$ and is analytically less convenient than (9). It is, of course, quite possible that the above formula, or some other formula, is much more accurate than (9) for the values of l where we need it, which are between 40 and 200, but we have found no evidence of this.

When it comes to the individual coefficients a_l and b_{l+1}, as opposed to their ratio, our information is less sure, since exact information about them is available only for $l \leqslant 5$. It can be argued, however,(14,22) (see also Appendix A) that for large l we have roughly

$$a_l \approx 4\pi D R_l \tag{11}$$

where D is the diffusion coefficient for monomers and R_l is the radius of an l-particle cluster (assumed spherical). If we accept the suggestion of Eq. (9) that the radius of curvature of such a cluster is proportional to $(l - 2)^{1/3}$, then we shall have

$$a_l = \alpha(l - 2)^{1/3} \tag{12}$$

where α is a constant. A value for α can be estimated by using the Einstein relation for the diffusion constant and by taking R_l to be the radius of a sphere of volume $l - 2$. The Einstein relation gives

$$D = \Delta x^2/(6\Delta t) = 1/12 \tag{13}$$

since for $\Delta t = 1$ our choice for the function F in Eq. (1) makes $\langle \Delta x^2 \rangle = F(0) = 1/2$. The estimated value for α is therefore

$$\alpha = 4\pi(1/12)(3/4\pi)^{1/3} = 0.65 \tag{14}$$

These approximations can now be used to help simplify the Becker–Döring equations. We first use (7) and the first equation of (2) to bring the Becker–Döring equations to the form

$$J_l = v_l c_l \tag{15}$$

where

$$v_l = a_l(1 - \rho)^3(w - w_l c_{l+1}/c_l) \tag{16}$$

Next, we use the approximations (9) and (12) for w_l and a_l to express v_l in the form

$$v_l = A\left\{\left(\frac{l - 2}{l^* - 2}\right)^{1/3} - 1 + \frac{w_l}{w_s}\left(1 - \frac{c_{l+1}}{c_l}\right)\frac{(l - 2)^{1/3}}{2.415}\right\} \tag{17}$$

where l^* is defined by

$$w = w_s[1 + 2.415/(l^* - 2)^{1/3}] \tag{18}$$

and A by

$$A = 2.415(1 - \rho)^3 a w_s \approx 0.013 \tag{19}$$

The variable l^* is our approximation to the size of a "critical" cluster over which the "vapor pressure" is equal to its actual instantaneous value w; that is, it is the size of a cluster that is as likely to grow as to shrink when the value of the "vapor pressure" is w. The value 0.013 for A is not reliable: in deriving it we used the extrapolation formula (9), we assumed, unrealistically, that clusters are spherical, and we used the Becker–Döring assumption that clusters of size 2 or greater never meet one another. A derivation that eliminates this second assumption, and gives a larger numerical value for A, is outlined in Appendix A.

Finally, as an approximation for the third-stage evolution of large clusters, we assume that, for large l, the scale of variation of c_l is l^*, so that $1 - c_{l+1}/c_l$ has the order of magnitude $1/l^*$. If we assume further that l^* itself is large, then we may approximate the formula (17) for v_l by

$$v_l \approx A[(l/l^*)^{1/3} - 1] \qquad (\text{large } l) \tag{20}$$

a formula due to Lifshitz and Slyozov.$^{(14)}$

In the same spirit, we may also approximate (4), for large l, by the partial differential equation

$$\frac{\partial c(l, t)}{\partial t} = -A \frac{\partial}{\partial l}\left[\left(\frac{l}{l^*}\right)^{1/3} - 1\right] c(l, t) \tag{21}$$

where l^* depends on w through the condition (9), and $c(l, t)$ is a smooth function of two continuous variables, so chosen that $c(l, t) = c_l(t)$ when l is an integer.

By itself Eq. (21) is not enough to determine $c(l, t)$ from a given initial distribution of clusters because it does not tell us how w and therefore l^* varies with time. To close the system of equations we use, as before, the particle conservation condition (6). Under the approximations we have been using, this condition takes the form

$$\rho_L(w) + \int_L^\infty c(l, t) l \, dl = \rho = \text{const} \tag{22}$$

where $\rho_L(w)$ is defined by

$$\rho_L(w) = (1 - \rho)^3 w + (1 - \rho)^4 \sum_{l=2}^{L} l w^l Q_l \tag{23}$$

and L is the point at which we choose to draw the line between "small" and "large" clusters. The value of L should satisfy the inequalities

$$1 \ll L < l^* \tag{24}$$

since L must be $\gg 1$ to justify the approximations leading to (21) and (22) and must be $< l^*$ because the quasiequilibrium approximation (2) does not apply to clusters larger than l^*: the minimum it predicts in c_l at $l = l^*$ is not in fact found.

Equations (20)–(21) are identical to the ones used by Lifshitz and Slyozov (LS). They treat l as a continuous variable from the beginning and do not distinguish between w and the density of the vapor. Equation (22) then corresponds to their simpler relation

$$w + \int_0^\infty c(l, t) l \, dl = \rho$$

The relation between w and l^* in the LS theory, with $w - w_s$ proportional to $(l^*)^{-1/3}$ instead of $(l^* - 2)^{1/3}$, is also simpler than ours, but to relative order $1/l^*$ it makes no difference. We believe that these differences between the LS equations and ours will not significantly affect the qualitative behavior of the solutions of these equations for large l. In later sections we discuss these solutions and compare them with our simulations.

4. SOLVING THE KINETIC EQUATIONS

It is argued by Lifshitz and Slyozov[(14)] that the kinetic equations for the large clusters, Eqs. (21) and (22), imply that l^* will increase linearly with time for large t. Table III shows a test of this prediction. Each value of t in this table is the average of 20 successive times at which "observations" of the cluster distribution (i.e., the values of the c_l) were made and recorded by the computer. Some of these values of t also appear in Table I. For each value of

Table III. Test of the Lifshitz–Slyozov Prediction that the Critical Cluster Size l^* Increases Linearly with Time

t	900	1189	1494	1796	2121	2454	2798	3137	3500	3870	4250	4640	5034	5431	5669
w/w_s	1.718	1.670	1.628	1.600	1.591	1.567	1.527	1.513	1.479	1.464	1.436	1.426	1.401	1.407	1.410
l^*	40.1	48.9	59	67.3	70.1	79.3	98.3	106.1	130.4	143.0	171.9	184.2	220.8	211.7	205.9
$0.0384t$	23	46	57	69	81	94	107	120	134	149	163	178	193	209	218

t shown, a value of w was calculated by the method described in connection with Table I. These values of w were used to calculate the values of w/w_s recorded in the second row of Table III. The values of l^* given in the third row were then calculated using Eq. (18), and finally the coefficient of t in the last row was calculated to give the least-squares best fit to the values of l^*. Comparison of the last two rows shows that the formula

$$l^* \approx 0.0384t \tag{25}$$

gives l^* with an accuracy of about 15%. This accuracy is not spectacular, but nevertheless is only about 3% of the total variation of l^* itself, and so confirms Lifshitz and Slyozov's prediction as a good first approximation to the truth.

To develop the consequences of Eq. (25), let us write, more generally,

$$l^* = Kt \tag{26}$$

The equation obtained by making this substitution in (21) can be solved using the method of characteristics after the further substitution

$$g(l, t) = \int_l^\infty c(\lambda, t)\, d\lambda \tag{27}$$

This new equation can be written

$$\frac{\partial g(l, t)}{\partial t} + \left[B\left(\frac{l}{t}\right)^{1/3} - A\right]\frac{\partial g(l, t)}{\partial l} = 0 \tag{28}$$

where

$$B = AK^{-1/3} \tag{29}$$

The characteristics of Eq. (28) are the solutions of

$$dl/dt = B(l/t)^{1/3} - A \tag{30}$$

that is,

$$\ln t + \phi(l/t) = \text{const} \tag{31}$$

where

$$\phi(x) = \int \frac{dx}{x - Bx^{1/3} + A} \tag{32}$$

The general solution of (28) is therefore

$$g(l, t) = \psi[\ln t + \phi(l/t)] \tag{33}$$

where ψ is an arbitrary function.

To use (33) (which is also due to Lifshitz and Slyozov) we write it in the form

$$\ln t = \psi^{-1}(g) - \phi(x) \tag{34}$$

where

$$x = l/t \tag{35}$$

and ψ^{-1} is the inverse of the function ψ. That is to say, the theory predicts that $\ln t$ is the sum of a function of $g(l, t)$, which we interpret as the number of clusters per lattice site larger than l at time t, plus a function of l/t. We obtained numerical information from the simulations about the functions ψ and ϕ by tabulating $\ln t$ as a function of the observed values of x and g. This is shown in Table IV. The columns are labeled by values of Ng rather than g, where $N = 125{,}000$ is the number of lattice sites, so that $Ng(l, t)$ is the number of clusters larger than l at time t. A sample of the data used in calculating Table IV is given in Appendix B.

In the main body of Table IV, each column corresponds to a value for $Ng(l, t)$, which is the number of clusters larger than l at time t, and each row corresponds to a value for $x = l/t$. The entries in the table were calculated in two stages from the data given in Appendix B (Table IX). First Ng, which is tabulated against l for various values of t in Table IX, was retabulated for those same values of t but at intervals of 0.01 in x, using linear interpolation. In the second stage, this new table was treated as a table of t against Ng and x, and retabulated using intervals of 5 in Ng using linear interpolation again. The logarithms of these interpolated values of t are the ones in Table IV.

If (34) is correct, then neighboring rows (columns) should differ by an amount which is the same in each column (row). The differences are shown in parentheses between the main entries. The table does approximately satisfy this condition, except in the rows for $x \geqslant 0.1$, i.e., $l \geqslant 0.10t \approx 2.6l^*$.

The $\Delta\phi$ column was calculated by averaging the entries in the relevant rows of differences; for example, the first entry 0.21 in this column is the average of the numbers 0.19, 0.21, 0.23 and 0.22. The ϕ column was then calculated by summing, after fixing $\phi(0.01)$ arbitrarily as 0.19, for example, $0.40 = 0.19 + 0.21$, $0.61 = 0.40 + 0.21$, etc. The ψ^{-1} row at the foot of the table was obtained by calculating the values of $\ln t + \phi(l/t)$ for the column of the main table above it and choosing an average or representative value. The $\Delta\psi^{-1}$ row shows the first differences of the ψ^{-1} row. In most cases the differences of neighboring entries in successive columns of the main table are close to the values given for $\Delta\psi^{-1}$.

All the tabulated values of $\ln t$, except for the five entries in the last three rows, satisfy Eq. (34) with an error not exceeding 0.05: that is, Eq. (34) gives t with an accuracy of 5% provided $x \leqslant 0.09$. Thus the table confirms the LS

Table IV. Computation of the Functions ϕ and ψ in Eq. (33)

	55	50	45	40	35	30	25	20	15	10	5	$\Delta\phi$	ϕ
0.01	—	—	—	7.70 (18)	7.88 (25)	8.13 (29)	8.42	—	—	—	—		0.19
				(19)	(21)	(23)	(22)					0.21	
0.02	7.15 (12)	7.27 (12)	7.39 (12)	7.51 (16)	7.67 (23)	7.90 (30)	8.20	—	—	—	—		0.40
	(24)	(25)	(24)	(18)	(16)	(20)	(22)					0.21	
0.03	6.91 (11)	7.02 (13)	7.15 (18)	7.33 (18)	7.51 (19)	7.70 (28)	7.98 (45)	8.43	—	—	—		0.61
			(28)	(28)	(25)	(22)	(25)	(30)				0.26	
0.04	—	—	6.87 (18)	7.05 (21)	7.26 (22)	7.48 (25)	7.73 (40)	8.13	—	—	—		0.87
				(24)	(25)	(25)	(18)	(10)				0.20	
0.05	—	—	—	6.81 (20)	7.01 (22)	7.23 (32)	7.55 (48)	8.03 (43)	8.46	—	—		1.07
						(22)	(26)	(32)	(27)			0.27	
0.06	—	—	—	—	—	7.01 (28)	7.29 (42)	7.71 (48)	8.19 (37)	8.56	—		1.34
							(27)	(25)	(32)	(26)		0.28	
0.07	—	—	—	—	—	—	7.02 (44)	7.96 (41)	7.87 (43)	8.30 (28)	8.58		1.62
								(38)	(28)	(26)	(25)	0.29	
0.08	—	—	—	—	—	—	—	7.08 (51)	7.59 (45)	8.04 (29)	8.33		1.91
									(33)	(35)	(29)	0.32	
0.09	—	—	—	—	—	—	—	—	7.26 (43)	7.69 (35)	8.04		2.23
										(23)	(10)	0.16	
0.10	—	—	—	—	—	—	—	—	—	7.46 (48)	7.94		2.39
										(31)	(13)	0.18	
0.11	—	—	—	—	—	—	—	—	—	7.15 (66)	7.81		2.57
											(22)	0.22	
0.12	—	—	—	—	—	—	—	—	—	—	7.59		2.79
$\Delta\psi^{-1}$	0.11	0.11	0.15	0.18	0.23	0.29	0.43	0.47	0.41	0.39			
ψ^{-1}	7.54	7.65	7.76	7.91	8.09	8.32	8.61	9.04	9.51	9.92	10.31		

prediction (34) about the functional form of $g(l, t)$. The method of analysis leading to this table, and from it to the functions ϕ and ψ, is independent of the extrapolation formula (9); it requires only that l^* be proportional to the time.

To carry this test of the Lifshitz–Slyozov theory further, we would like to have approximate formulas for the functions ϕ and ψ having some basis in theory. We start with the function ϕ, since, according to Eq. (32), two parameters A and B are sufficient to fix this function completely. Since (32) implies that the derivative ϕ' of the function ϕ satisfies

$$[1/\phi'(x)] - x = A - Bx^{1/3} \tag{36}$$

we can find values for A and B by plotting $[1/\phi'(x)] - x$ against $x^{1/3}$ and fitting a straight line. Using the values $x = 0.015, 0.025, \ldots$ and estimating the corresponding values of $\phi'(x)$ from the finite differences $\Delta\phi$ given in Table IV, we found the least-squares best fit to be given by

$$A = 0.1525, \qquad B = 0.4530 \tag{37}$$

When combined with (29) this gives $K = 0.0382$, so that (26) becomes $l^* = 0.0382t$. This agrees very well with the value 0.0384 given in (25), which was obtained by an independent method, and so provides some confirmation of the ideas underlying our method of analysis. On the other hand, the above value for A is over ten times as big as the first-principles estimate given in (19). Some reasons for mistrusting that estimate were mentioned just after (19). Our results also deviate at this point from a prediction of Lifshitz and Slyozov, that the function $1/\phi'(x)$ must have a double zero. The condition for a double zero is $A/K = 27/4 = 6.75$, whereas we find $A/K = 3.99$, which implies that the expression (36) for $1/\phi'(x)$ has no zeros at all for positive values of x. A brief discussion of the value of A/K, that is, B^3/A^2, is given at the end of this paper.

Although the values of A and B given in (35) gave the best fit to the values of $[1/\phi'(x)] - x$, it turned out that the values of $\phi(x)$ itself, obtained in Table III, could be fitted better by taking $A = 0.154$ instead of 0.1525. This value for A, with $B = 0.453$ as before, implies $K = 0.0393$, still remarkably close to the value 0.0384 given in (25). Table V shows a comparison of the values of $\phi(x)$ from Table IV with those implied for formula (36) with $A = 0.154$, $B = 0.453$, and a constant of integration chosen to give the best agreement [the value of this constant of integration is such that $\phi(0) = 0.09$].

To obtain an analytical formula for the function ψ, we use the identity (22), which, when combined with (27), gives

$$\rho - \rho_L(w) = \int_L^\infty \left(-\frac{\partial g(l, t)}{\partial l}\right) l\, dl = Lg(L, t) + \int_L^\infty g(l, t)\, dl \tag{38}$$

Table V. Values of $\phi(x)$ from Table IV Compared with the Formula (36)

x	0.01	0.02	0.03	0.04	0.05	0.06
$\phi(x)$	0.19	0.40	0.61	0.87	1.07	1.34
Formula	0.20	0.37	0.59	0.83	1.10	1.37

x	0.07	0.08	0.09	0.10	0.11	0.12
$\phi(x)$	1.62	1.91	2.23	2.39	2.57	2.79
Formula	1.64	1.90	2.15	2.39	2.61	2.82

From this and our formula (33), which we are supposing to give $g(l, t)$ when $l \geqslant L$, it follows that

$$\int_0^\infty \psi[\ln t + \phi(l/t)]\, dl$$

$$= \rho - \rho_L(w) - \int_0^L \{\psi[\ln t + \phi(L/t)] - \psi[\ln t + \phi(l/t)]\}\, dl \qquad (39)$$

Assuming that the functions ψ and ϕ have bounded first derivatives, the integral on the right side of (39) has an upper bound of the form const/t. The expression $\rho - \rho_L(w)$, on the other hand, approaches the constant $\rho - \rho_L(w_s)$ for large t. Lifshitz and Slyozov argue from this that the integral on the left is approximately independent of t, and hence that $\psi(x)$ is approximately proportional to e^{-x}. At the times considered in our simulations, however, this approximation is not a good one; the reason is that even at the end of our run w and $\rho_L(w)$ were still far from their asymptotic values. For a very large system these asymptotic values would be $w_s = 0.0105$ and $\rho_L(w_s) = 0.0118$, whereas the values at the end of our run were $w_s = 0.0148$ and $\rho_L(w_s) = 0.0213$. It is true that the asymptotic values of w and $\rho_L(w)$ are somewhat larger for our system because it is of finite size—we estimate the asymptotic values to be $w = 0.012$ and $\rho_L(w) = 0.015$—but the simulation was still far from these values when it ended.

To obtain a better approximation for ψ we use the formulas (18) and (25), which imply

$$w \approx w_s(1 + 7.17/t^{1/3}) \qquad (40)$$

To obtain from this a formula for $\rho_L(w)$ as a function of t, we fitted a quadratic polynomial to the function $\rho_L(w)$ for the relevant range of values of w, which we took to be $w_s \leqslant w \leqslant 1.7w_s$, since w was approximately $1.7w_s$ at the earliest time considered in these calculations. We tried this method both for $L = 10$ and for $L = 20$. The results were more convincing for $L = 20$, since

Table VI. Values of $\rho_{20}(w)$ from Eq. (2) Compared with the Quadratic Approximation (41)[a]

w/w_s	1.0	1.1	1.2	1.3	1.4	1.5	1.6	1.7
$125{,}000\rho_{20}(w)$	1480	1715	1984	2295	2669	3134	3745	4605
Quadratic	1480	1668	1936	2284	2713	3221	3810	4478

[a] The values of ρ_{20} in this table were calculated from (2), using (8) and (9) to get $Q_{11},\ldots, Q_{20}$.

the empirical distribution formula (2) predicts a nonnegligible number of clusters of sizes between 11 and 20 at the higher values of w considered. The quadratic approximation used for $L = 20$ was

$$125{,}000\rho_{20}(w) \approx 1480 + 481y + 423y^2 \tag{41}$$

where $y = (40/13)[(w/w_s) - 1]$. Table VI shows that the accuracy of this approximation is roughly 3%.

When (40) is substituted into (41) we obtain

$$125{,}000[\rho - \rho_{20}(w)] \approx 7895 - 10{,}612t^{-1/3} - 205{,}879t^{-2/3}$$

Substituting this into (39) and neglecting the integral on the right, we obtain

$$\psi(x) = C_0e^{-x} - C_1e^{-4x/3} - C_2e^{-5x/3} \tag{42}$$

where

$$\begin{aligned} NC_0 &= 7895\Big/\int_0^\infty e^{-\phi(x)}\,dx \\ NC_1 &= 10{,}612\Big/\int_0^\infty e^{-4\phi(x)/3}\,dx \\ NC_2 &= 205{,}879\Big/\int_0^\infty e^{-5\phi(x)/3}\,dx \end{aligned} \tag{43}$$

with $N = 125{,}000$.

Approximate values of the integrals were obtained from the values of ϕ given in Table IV, using Simpson's rule for $0 \leqslant x \leqslant 0.12$ and a linear extrapolation of the function ϕ to estimate the contribution of values of x exceeding 0.12. These values, substituted into (43), gave

$$\begin{aligned} NC_0 &= 7895/0.0449 = 1.76 \times 10^5 \\ NC_1 &= 10{,}612/0.0341 = 3.11 \times 10^5 \\ NC_2 &= 205{,}879/0.0276 = 7.46 \times 10^6 \end{aligned} \tag{44}$$

Table VII. Values of $N\psi(x)$ from Table IV Compared with the Formula (42)

	x	7.54	7.65	7.76	7.91	8.09	8.32	8.61	9.04	9.51	9.92	10.31
$N\psi(x)$:	from (42)	54.1	50.6	47.1	42.4	37.1	31.1	24.5	16.9	11.1	7.6	5.3
	from Table IV	55	50	45	40	35	30	25	20	15	10	5

Table VII shows the values of $\psi(x)$ calculated from (42) with these coefficients, compared with the "observed" values taken from Table IV. (The symbol x has different meanings in the two tables.)

5. CONCLUSIONS AND DISCUSSION

The main conclusions to be drawn from these computer simulations and our analysis of them are these:

1. Over the time range considered (which started fairly late in the process, when there had already been about 1000 attempted interchanges per site) the distribution of small clusters is given approximately by the empirical formula (2), with a value of w that changes slowly with time.

2. The time dependence of w can be approximately represented by the formula

$$w \approx w_s[1 + 2.4/(l^*)^{1/3}] \tag{45}$$

where $w_s = 0.0105$ is the value of w describing the saturated vapor, and l^* is given by

$$l^* \approx 0.038t \tag{46}$$

with t the time, measured by the number of attempted interchanges per site. The fact that l^* is proportional to t accords with the predictions of Lifshitz and Slyozov.(14)

3. The distribution of large clusters is described by a formula of the type implied by the theory of Lifshitz and Slyozov(14)

$$c(l, t) = \frac{d}{dl}\psi\left\{\ln t + \phi\left(\frac{l}{t}\right)\right\} \tag{47}$$

where

$$\phi(x) = \int \frac{dx}{x - Bx^{1/3} + A} \tag{48}$$

and the function ψ can be determined by considering the total number of particles in large clusters.

4. It is a consequence of (47) that the average size of the large clusters, which is roughly l^*, grows in proportion to t. Various investigators, including some of the present authors,(23),[8] have reported average cluster sizes growing more slowly than this (e.g., as $t^{3/5}$). The reason for the discrepancy may be that the times considered were too early, or the concentrations too high, for the LS theory to apply; or that the averages included some small clusters as well as the large ones.

5. The function ψ is fitted quite well by the formula

$$\psi(x) = C_0 e^{-x} - C_1 e^{-4x/3} - C_2 e^{-5x/3} \tag{49}$$

with C_0, C_1, C_2 determined by considering how w, and hence the number of particles in large clusters, changes with time. In the very late stages of the process, to which the Lifshitz–Slyozov theory applies, only the term $C_0 e^{-x}$ is necessary, but for the times considered in our simulations the other terms are necessary, too.

6. For the normalization integral associated with the formula (49) to converge, it is necessary that $B^3/A^2 \geqslant 6.74$, as noted by Lifshitz and Slyozov.(14) The observed value of B^3/A^2 was 4.0. This seems to indicate that our results cannot be extrapolated to arbitrarily large values of $\ln t + \phi(l/t)$. Possibly the value of B eventually gets larger than the value 0.453 which we found. This would imply, by (29), a smaller value of $K = (A/B)^3$, that is, a smaller rate of increase of l^*, than during the time interval covered by our simulations. To be consistent with the condition $B^3/A^2 \geqslant 6.75$, this "ultimate" value of K would have to be not greater than $A/6.75 = 0.0226$, instead of the value 0.038 implied by our data.

APPENDIX A. CLUSTER GROWTH BY DIFFUSION

In the body of this paper we used a formula (11) relating the rate at which particles arrive at the surface of an l-particle cluster to the diffusion coefficient for monomers. In the derivation of that equation(14,22) it is assumed that almost all the small clusters are monomers. For the densities we are considering here, a sizeable fraction of the small clusters are not monomers, and so an improvement on formula (11) and its consequence, Eq. (19), is desirable. To provide this we consider the process of diffusion near a cluster of size l without making any specific assumptions about the other clusters. Consider two neighboring lattice sites, say site 1 and site 2, in a nonuniform system for which the probability of site i being occupied (by

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[8] See Ref. 8 for other references.

an A particle, in alloy language) is ρ_i. The probability of the event that site 1 is occupied and site 2 empty (occupied by a B particle, in alloy language) is $\rho_1(1 - \rho_2) + O(\rho^3)$, and the conditional probability, given this event, that the particle (A particle) moves from site 1 to site 2 is (in an approximation which becomes exact as $\rho \to 0$) $\frac{1}{6}F(0)$ per unit time, where F is defined in Eq. (1) and was put equal to 1/2 in our simulation. The probability that the (A) particle will jump from site 1 to site 2 is therefore approximately $\frac{1}{6}F(0)\rho_1(1 - \rho_2)$ per unit time. Similarly the probability of a jump from site 2 to site 1 is $\frac{1}{6}F(0)\rho_2(1 - \rho_1)$. Subtracting, we find that the mean rate of flow from site 1 to site 2 is

$$\tfrac{1}{6}F(0)[\rho_1(1 - \rho_2) - \rho_2(1 - \rho_1)] = \tfrac{1}{12}(\rho_1 - \rho_2)$$

It follows that the density of (A) particles satisfies a lattice version of the diffusion equation with diffusion constant $D = 1/12$.

The quantity v_l defined in Eq. (15) is the average net number of particles per second arriving at the surface of an l-particle cluster, which we take to be a sphere of radius R_l. On the assumption that diffusion controls the rate of arrival and absorption of these particles, the density $\rho(r)$ at distance r from the center of the cluster is related to the radial diffusion current $I(r)$ by

$$I(r) = -D\, d\rho(r)/dr \tag{A1}$$

where D is the diffusion constant. Assuming a steady state ($v_l = \text{const}$) the conservation of particles requires

$$4\pi r^2 I(r) = \text{const} = v_l \tag{A2}$$

Using this in (A1), with the boundary condition $\rho(r) \to \rho$ as $r \to \infty$, we find

$$\rho(r) = \rho - (v_l/4\pi D r)$$

The boundary condition at $r = R_l$ then gives

$$v_l/4\pi D R_l = \rho - \rho(w_l) = (w_{l*} - w)\, d\rho/dw + \cdots$$

where we are assuming that it is possible to associate a density $\rho(w)$ with each value of w. Substituting from (9) for w_l, (13) for D, and from the assumed relation $l - 2 = (4\pi/3)R_l^3$, as in the text, for R_l, we obtain Eq. (20), but with A now given by

$$A = 4\pi \frac{1}{12}\left(\frac{3}{4\pi}\right)^{1/3} 2.415 \frac{d\rho}{dw}$$

The main difference between this formula and the formula (19) for A given in the text is the factor $d\rho/dw$, which allows for the contribution of clusters containing more than one particle to the supply of particles to the cluster of size l. The numerical value of $d\rho/dw$ depends on what function we assume for $\rho(w)$. A reasonable choice, based on the assumption that (2) is valid for all $l \leqslant l^*(w)$, would be

$$\rho(w) = (1 - \rho)^3 w + (1 - \rho)^4 \sum_2^{l^*} l Q_l w^l$$

but for simplicity we truncated the series at $l = 20$ instead of $l = l^*$. Our procedure gives a value of $d\rho/dw$ which decreases from about 3 to 1.5 as the run considered in this paper proceeds. This extra factor in the theoretical formula for A brings up the value of A from 0.013 [as obtained in Eq. (19)] to a value in the range 0.02–0.04. Unfortunately, the observed value of A given in Eq. (37) is still considerably larger.

APPENDIX B. SOME OF THE DATA

Table VIII shows a sample of the data used in the investigation. In each row of the table the left-hand entry is a time and the other numbers are the sizes of the 25 largest clusters at that time. The numbers in the 20 rows between the two horizontal rules are among those used in calculating the averaged values of $Ng(l, t)$ for $t = 2121$, which were then combined with averaged values of $Ng(l, t)$ for the other times in Table III to give the entries in Table IV.

Table VIII illustrates how the size of the largest cluster can be affected by coagulation of other large clusters. The largest cluster at time 1980.6 has size 285, but by time 2013.5 this cluster (now of size 278) is only second largest; the new largest cluster, of size 441, appears to have been formed by the coagulation of two of the five clusters whose sizes at time 1980.6 were between 214 and 228. A similar event can be seen between times 1786.7 and 1802.6, but this one is quickly undone again.

Table IX shows the values of $Ng(l, t)$ used in compiling Table IV. The numbers at the heads of the columns are time t. Of the remaining numbers, those on the right-hand side of each column are equally spaced values of l; those on the left are the corresponding values of $Ng(l, t)$, the number of clusters larger than l, averaged over 20 times whose average is t.

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Table VIII. Sizes of the 25 Largest Clusters at a Succession of Times

1802.6	407	265	241	238	234	224	219	214	130	176	172	165	161	154	148	146	134	121	120	118	116	111	105	104	103
1818.5	414	246	241	233	232	228	227	218	210	176	176	174	172	166	155	155	146	142	131	131	120	113	104	104	98
1834.6	247	236	235	234	233	226	219	211	209	185	176	170	168	164	156	152	143	132	129	128	124	121	117	102	101
1850.6	255	240	239	236	236	225	213	212	204	188	179	177	164	163	161	154	138	137	130	125	120	119	113	111	103
1866.7	257	242	241	235	233	224	223	211	210	205	185	183	174	165	163	157	147	140	133	130	128	115	108	102	102
1882.8	266	244	241	240	222	219	215	209	208	184	182	177	169	165	159	151	150	141	138	130	128	127	122	111	102
1899.0	265	252	234	227	223	221	219	216	211	181	179	177	171	167	154	153	148	142	138	128	125	123	119	113	103
1915.2	295	251	240	233	228	221	220	213	195	187	177	172	170	168	160	158	150	143	132	130	127	127	119	118	105
1931.5	282	249	241	234	229	229	215	214	212	179	174	169	169	162	156	156	149	139	133	129	129	127	118	113	112
1947.9	274	245	243	241	234	224	220	215	209	184	181	174	172	153	153	151	148	148	139	133	127	123	123	117	101
1964.2	282	250	249	238	219	219	217	214	213	191	179	172	168	156	155	152	152	151	139	134	130	124	123	120	111
1980.6	285	253	244	230	228	223	220	216	214	190	183	175	172	170	162	161	157	146	138	133	127	126	126	119	110
1996.9	441	278	250	247	238	225	218	218	197	182	171	171	169	165	163	157	145	141	140	133	130	125	121	111	102
2013.5	432	271	259	252	243	230	224	215	199	191	179	176	169	168	167	164	151	144	141	135	133	129	120	102	102
2029.9	434	284	268	257	251	230	226	217	203	190	179	178	171	168	167	165	156	146	143	140	127	125	115	101	97
2046.5	414	293	258	251	247	231	230	218	202	192	189	184	179	169	165	165	164	140	132	128	128	126	125	107	88
2063.0	345	291	275	262	254	240	224	214	210	203	203	202	187	177	173	169	168	160	134	130	128	128	128	105	90
2079.6	411	290	274	246	244	233	220	218	209	203	182	175	173	172	166	165	157	140	136	130	126	126	122	106	91
2096.0	423	339	284	277	245	241	227	223	221	212	182	178	173	172	170	170	155	142	137	130	128	115	106	100	92
2112.7	417	337	284	283	249	244	235	220	216	204	178	178	178	177	171	169	157	148	141	131	118	113	112	105	88

2129.4	424	342	288	277	240	239	238	220	218	204	182	180	180	177	169	162	149	140	133	130	113	113	109	94	92
2145.8	431	297	276	247	238	232	225	211	205	203	187	180	179	177	173	167	165	152	152	143	132	128	123	114	114
2162.4	420	307	304	297	241	239	229	227	222	218	200	181	181	175	173	158	153	141	138	126	120	118	114	109	103
2178.9	416	337	306	293	250	246	236	231	218	218	183	177	176	174	171	160	148	147	141	132	124	113	112	109	97
2195.4	402	344	290	288	250	242	237	226	223	217	197	186	174	172	166	157	152	148	143	127	119	117	111	102	97
2211.8	417	320	300	295	247	244	243	226	216	212	183	183	176	173	169	161	149	140	138	136	125	108	108	105	101
2228.1	417	331	293	287	261	252	251	215	211	203	194	183	178	174	165	164	164	141	140	137	127	109	109	100	96
2244.4	422	330	299	295	249	248	240	219	209	204	198	194	186	178	169	161	147	144	141	140	134	105	98	94	92
2260.7	416	334	301	290	250	247	240	220	218	212	192	190	185	175	175	164	155	146	146	142	123	98	98	93	92
2277.1	467	342	301	286	257	250	249	224	217	214	203	189	177	176	173	165	147	144	142	140	120	107	94	93	93
2293.8	415	340	301	294	291	260	251	236	226	221	208	195	190	176	175	157	153	149	144	141	124	102	98	95	87
2310.4	411	344	320	300	270	253	239	226	225	205	199	196	186	184	180	165	155	148	144	143	117	102	98	96	89
2327.2	415	347	318	294	276	255	236	224	210	209	206	195	192	188	188	173	160	153	144	143	123	120	97	94	92
2344.0	475	346	317	295	293	286	246	226	217	215	210	200	195	191	190	177	153	152	143	140	130	110	99	95	93
2361.2	437	342	313	304	291	280	250	232	220	210	205	192	191	188	179	179	161	151	148	145	133	116	104	100	96
2378.2	432	319	315	301	256	239	236	233	213	210	207	207	204	195	179	171	160	156	153	145	135	129	117	111	94
2395.1	431	314	307	301	256	247	236	220	212	211	207	202	200	194	183	176	159	157	146	144	136	127	116	109	93
2411.9	437	363	305	300	300	256	248	236	231	223	210	206	205	197	183	176	164	152	150	137	131	111	110	88	88
2428.8	435	308	308	256	253	242	238	227	219	209	208	208	204	187	187	187	169	152	152	143	139	129	110	107	99
2445.8	430	318	316	302	265	259	246	229	221	208	208	203	199	199	190	187	170	153	148	145	144	116	114	110	94
2462.7	451	320	312	299	256	244	243	229	222	219	211	205	199	197	193	190	160	156	150	141	141	119	116	106	103

Table IX. Values of *Ng(l, t)* at Various Times *t* for Different Sizes *l*

900	1189	1494	1796	2121	2454	2798	3137	3500	3870	4250	4690	5034	5431
72 20	61 20	57 20	51 20	42 20	38 20	33 20	33 20	31 20	28 20	28 20	25 20	25 20	25 20
57 29	50 32	49 28	42 32	37 34	34 37	33 36	32 36	29 38	27 40	26 40	25 43	25 43	24 43
45 37	41 44	45 36	38 45	34 48	31 55	30 51	28 51	27 56	26 59	25 61	23 65	23 66	23 66
40 46	35 56	41 44	34 57	31 63	27 72	27 67	27 67	26 74	23 79	23 81	22 88	22 89	22 89
35 54	29 68	37 52	30 70	28 77	25 89	26 82	26 83	24 92	22 98	22 101	22 111	21 118	20 112
29 63	25 70	33 60	27 82	25 91	22 107	24 98	23 99	21 110	21 118	22 122	20 134	20 135	20 135
24 71	21 92	30 68	25 95	23 105	21 124	22 113	22 114	21 128	20 137	21 142	19 156	19 158	19 158
19 80	18 104	28 76	23 107	22 120	20 142	21 129	21 130	19 146	18 157	19 163	18 179	18 181	18 181
15 88	15 115	26 85	21 120	19 134	17 159	20 144	20 146	19 163	18 177	19 183	18 202	18 203	17 203
12 97	12 127	24 93	18 132	17 148	16 176	19 160	20 162	18 181	17 196	18 203	17 224	17 226	16 226
10 105	8 139	22 101	16 145	16 162	13 194	17 175	18 177	17 199	15 216	15 224	14 247	13 249	12 249
9 114	6 151	20 109	14 157	12 177	10 211	14 191	15 193	15 217	14 235	13 244	11 270	11 272	11 272
6 122	5 163	19 117	12 170	10 191	8 228	13 306	15 209	13 235	13 255	12 264	10 292	11 295	11 295
5 131	5 175	17 125	9 182	9 205	6 246	11 222	13 225	12 253	11 274	9 285	9 315	9 318	8 318
4 139	4 187	15 133	8 195	7 219	5 263	10 237	11 240	9 271	9 294	7 305	6 338	7 341	7 341
3 148	4 199	13 141	8 207	6 234	4 280	8 253	9 256	6 289	6 314	5 326	5 360	5 364	6 364
3 157	2 211	11 149	6 219	4 248	4 298	6 269	6 272	5 307	5 333	4 346	4 383	4 387	4 387
2 165	2 223	9 157	4 232	3 262	3 315	4 284	4 287	4 325	4 353	4 366	2 406	2 410	3 410
2 174	1 235	8 165	1 244	3 276	1 333	4 300	4 303	4 343	3 372	3 387	2 429	2 433	2 433
1 182	1 247	7 173	1 257	2 290	1 350	4 315	4 319	3 361	2 392	2 407	1 451	2 456	2 456
1 191	1 259	6 181	0 269	2 305	1 367	3 331	3 335	2 279	2 411	1 427	1 474	1 479	2 479
1 199	1 271	6 189		1 319	1 385	2 346	2 350	2 397	2 431	1 447	1 497	1 502	1 502
1 208	1 283	5 197		1 333	1 402	2 362	1 366	2 415	1 450	1 468	1 519	1 525	1 525
1 216	0 294	5 206		1 347	1 419	2 377	1 382	2 432	1 470	1 488	1 542	1 547	1 547
1 225	0 306	4 214		1 362	1 437	1 393	1 398	1 450	1 490	1 509	1 565	1 570	1 570
1 233	0 318	3 222		1 376	0 454	1 408	1 413	1 468	1 509	1 529	1 588	1 593	1 593
1 242	0 330	2 230		1 390		1 424	1 429	1 486	1 529	1 550	1 610	1 616	1 616
1 250	0 342	1 238		1 404		1 439	1 445	1 504	1 548	1 570	1 633	1 639	1 639
0 259	0 354	1 246		0 419		1 455	1 461	1 522	1 568	1 590	0 656	1 662	1 662
	0 366	0 254				1 470	1 476	0 540	1 587	1 611		0 685	1 685

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