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Nucleation in Condensed Matter

Applications in Materials and Biology

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NUCLEATION IN CONDENSED MATTER

APPLICATIONS IN MATERIALS AND BIOLOGY

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This book is dedicated to the memory of

DAVID TURNBULL
(1915–2007)

for his leadership in research, directing the authors towards the study of nucleation

ROBERT CAHN
(1924–2007)

for his guidance and encouragement, driving onward the writing of this book

PREFACE

Nucleation, for example involving the spontaneous appearance of small regions of a new phase in an old phase, is the usual starting process for transformations in a material. While the study of nucleation is an old one, stretching back to pioneering work by Fahrenheit in the early 1700's, it remains a very active field, on which several books have just recently appeared. Some overlap of the material covered in this book with that covered in those is unavoidable. However, we hope that our presentation is distinctive in covering a wide range of topics, and in taking the chosen topics up to the frontier of current work. The key theories of nucleation are presented and critiqued in light of experimental tests. Coupled processes that often underlie nucleation are discussed, as is the important practical theme of nucleation control. Given the number of unanswered questions in the field and the rapid progress being made, some sections of this book will soon appear dated; we feel, nevertheless, that it is useful to have a survey of the present state of the art. We have tried to provide the reader with comprehensive citations of original sources. This has involved much effort, starting from reference data that were often patchy or wrong. We now have close sympathy with the editors of the *Catalogue of Scientific Papers 1800–1863*, published by the Royal Society of London in 1867, who bemoaned the lack of complete data in tracing authors and publications: "None but those who have been engaged in a task of this kind can form any idea of the difficulty occasioned by such omissions."

This book aims to provide a comprehensive coverage of nucleation in condensed matter, and includes some topics (such as nucleation in biology, medicine, food and drink) that fall outside the usual focus in materials science and physics. We are, however, conscious of such omissions as the nucleation of magnetic and other ordered domains, and nucleation in vapors, which has practical application in atmospheric processes that are of increasing environmental concern. A discussion of these and other topics was originally planned, but it became clear that their inclusion would increase the length of this book beyond a reasonable limit. The study of nucleation in vapors, for example, could easily be the focus of another book of similar length. Inevitably, the choice of topics reflects the background and prejudices of the authors, whose own research has focused on the nucleation of crystals in liquids and glasses, but we hope that other important topics are fairly represented.

Having completed the book, we are struck by the evident lack of communication even within the field of nucleation studies. To cite just one example of many: line tension has been well studied for heterogeneous nucleation of condensation from a vapor, but has been largely ignored for crystallization of a liquid. We hope that in many ways, not least bridging such communication gaps, this book can stimulate further advances in nucleation studies.

It is a pleasure to dedicate the book to two particularly distinguished scholars. A glance even at the name index of this book will show that the influence of David Turnbull on studies of nucleation in condensed matter was pervasive and continues to be so. We both had the privilege of working in Turnbull's laboratory at Harvard, and our scientific collaboration stems from that time. In some sense this is the book that we wish Turnbull had written. Although he might have done it better, we have been able to take the story further on in time. This book would never have come about had it not been for the continual encouragement of Robert Cahn in the other Cambridge, acting principally in his capacity as inaugural editor of the *Pergamon Materials Series*. His influence on us, by his own example, in the art of good writing has been immense, even if we fail to reach his standards.

In our research and in writing this book, we have benefited from interactions with many colleagues around the world. We thank them for discussion and their shared insights into many of the new ideas that are expressed here. ALG thanks the many graduate students who contributed to his education on nucleation while they worked in his group. We thank Professors Lev Gelb and Michael Ogilvie, Washington University, for a critical reading of Chapters 4 and 10, and Linda Coffin, Emily Kelton, and many of KFK's graduate students, for their editorial assistance and comments on chapters at various stages. KFK particularly thanks his wife, Emily, and sons Franklin and James for their support during the many years spent in the preparation of this book. Of course, despite all of the input from others, the responsibility for any errors and omissions lies with the authors. The companion site for this work: http://www.elsevierdirect.com/companion.jsp?ISBN=9780080421476 includes color versions of some of the figures and an updated sheet of errata.

Ken Kelton
Lindsay Greer
October 2009

SYMBOLS

Chapter and section numbers are cited where a symbol has different meanings in different sections.

Symbol	Definition
	Prefixes
δ	incremental change
Δ	change or difference
	Subscripts
0	initial value
am	austenite/martensite
amor	amorphous
appl	applied
A	atoms; Avogadro
b	bubble
bu	break-up
B	Boltzmann, boundary, Bragg
c	coagulation; critical; crystallization (of a single polymer chain)
cap	spherical cap
cf	composition fluctuation; coupled-flux
ch	ccp/hcp
cl	cluster
class	classical
clus	cluster
col	columnar
core	dislocation core
crys	crystalline nucleus
ct	center
cyl	cylindrical
C	Curie
CNT	classical nucleation theory
d	droplet
disk	disk
D	free diffusion on lattice
DF	density-functional
DIT	diffuse-interface theory

eff	effective
emit	dislocation emission
eq	equilibrium
es	electrostatic
EM	electromigration
f	fusion
fg	free-growth
flu	fluctuation (of order or composition)
g	gas
gb	grain-boundary
gr	gram-atomic
grad	in a composition gradient
GLCH	Ginzburg-Landau Cahn-Hilliard
het	heterogeneous
hom	homogeneous
H	atomic hydrogen
He	helium
i	interfacial
inc	incubation
int	interstitial
ion	ion
IA	interfacial attachment
K	Kashchiev
Kn	Kauzmann
ℓ	dislocation line
l	liquid
lat	lateral (ledge movement on a crystal surface)
lf	linked-flux
lim	limiting
liq	liquidus
loop	dislocation loop
LJ	Lennard-Jones
m	medium (initial phase); melting
max	maximum
min	minimum
M	magnetic, molar
MWDA	modified weighted-density approximation
N	nucleation, nucleant phase
NC	nonclassical
Nuc	nucleant substrate or area
O	oxygen
pore	pore or bubble
pt	thermal plateau

P	nucleant patch; Poisson
PDFA	perturbative density-functional approximation
q	value to which a sample is quenched
r	reduced (i.e. normalized)
rel	relaxation
rept	reptation
res	resolved
s	shear; solid; start; first stem (of a polymer chain); supersaturation
sol	solidification; solutal
st	steady-state
step	surface step
strain	strain
sup	supersaturation
S	surface, per unit area
SCCT	self-consistent classical theory
SDFA	semi-empirical density-functional approximation
SF	stacking fault
th	thermal
us	unstable stacking
v	vapor
vac	vacancy
void	void
V	per unit volume
VW	Volmer-Weber
w	critical condition for wetting
WD	work done
α	original phase (α)
$\alpha\gamma$	in α phase in contact with γ phase; pertaining to the interface between α and γ phases
β	new phase (β)
γ	product phase (precipitate or interfacial reaction product)
κ	dielectric
	Superscripts
0	initial
+	forward
−	backward
′	final value
*	critical value (corresponding to the unstable equilibrium of a critical nucleus)
at	per atom
A	of A atoms

B	of B atoms
crit	critical
d	droplet
eq	equilibrium
l	liquid
m	medium (initial phase)
s	solid
st	steady-state
WDA	weighted-density approximation
z	for zero flux
	Main Symbols
a	geometrical factor [Ch. 2, §8] ∣ activity [Ch. 2, §11] ∣ the ratio $\Delta g_{sl}/\Delta g_{il}$ [Ch. 4, §3] ∣ capillary length ($= (2\sigma/k_B T)\bar{v}C_\infty$) [Ch. 4, §5] ∣ number of atoms/molecules of species A [Ch. 5, §1; Ch. 7, §2] ∣ lattice parameter, interatomic spacing [Ch. 4, §4; Ch. 9, §2; Ch. 14, §5; Ch. 15, §3; Ch. 16, §4]
a'	geometrical factor including the entropy of fusion per unit volume ($a' = a\, \Delta s_f^{-2}$) [Ch. 2, §8; Ch. 16, §4]
A	interfacial area [Ch. 2, §2] ∣ portion of A^* that is independent of the atomic mobility [Ch. 9, §2]
A	chemical species A
A	diagonalized matrix of **K**
A^*	kinetic (dynamical) pre-factor for nucleation rate (usually $\text{mol}^{-1}\,\text{s}^{-1}$)
$A(n)$	drift coefficient, $(k^-(n) - k^+(n))$ [Ch. 2, §10]
b	elements of matrix that diagonalizes **K** [Ch. 3, §3] ∣ $\sqrt{1-a}$, with a as in Ch. 4, Eq. (15) ∣ length of Burgers vector of dislocation [Ch. 6, §3; Ch. 12, §2; Ch. 14, §5] ∣ number of atoms/molecules of species B [Ch. 5, §1; Ch. 7, §2] ∣ thickness of a layer of chains (polymer crystal) [Ch. 11, §6] ∣ lattice parameter [Ch. 16, §4]
$\boldsymbol{b}$	Burgers vector
B	chemical species B
B	matrix of elements b diagonalizing **K**
B	applied magnetic field [Ch. 10, §3]
$B(n)$	drift coefficient, $\frac{1}{2}(k^-(n) + k^+(n))$ [Ch. 2, §10]
B_j	coefficient describing the interaction of j molecules [Ch. 4, §2]
B_J	the Brillouin function [Ch. 7, Eq. (21)]
c_p	specific heat at constant pressure
C	constant

C	solute concentration: C_∞ equilibrium C at the surface of an infinitely large particle; $\bar{C}$ average C; C_α^{i} concentration of solute in the α phase next to a precipitate; $C_{\gamma\alpha}$ concentration of A atoms in the product phase γ in contact with phase α
d	dimensionality of system [Ch. 4, §5] | average grain diameter in a polycrystalline phase [Ch. 6, §2; Ch. 13, §5]
D	diffusion coefficient ($\mathrm{m^2\,s^{-1}}$): D' effective D in size (n) space; $\tilde{D}$ interdiffusivity; D_{A}^* tracer diffusivity of A atoms
e	charge on an electron (1.602192×10^{-19} C)
e	natural logarithm base (2.71828)
Δe	elastic strain energy per unit volume [Ch. 14, §3]
E	applied electric field [Ch. 6, §4; Ch. 15, §5] | Young modulus [Ch. 9, §2] | energy: E_{ch} strain energy (per mole) associated with the ccp-to-hcp transformation [Ch. 12, §4]; E_{loop} total line energy of a dislocation half-loop [Ch. 12, §2]; E_n energy of the n^{th} state of the system [Ch. 10, §2]; E_{step} total energy of a surface step on a thin film [Ch. 12, §2]; E_{strain} uniform strain energy of a thin film [Ch. 12, §2]
E_1	single atom or molecule: E_n cluster of n atoms or molecules
f	Helmholtz free energy per unit volume: f_0 for a uniform system [Ch. 4, §4] | catalytic factor for heterogeneous nucleation: $f(\phi)$ as a function of contact angle ϕ; $f(\phi, X)$ as a function of contact angle ϕ and normalized nucleant size X [Ch. 6, §2] | order parameter: f_{bcc} for bcc; f_{ccp} for ccp; f_{liq} for the liquid [Ch. 10, §5]
F	Helmholtz free energy (usually per mole) [Ch. 2, §2]: F_{ex} particle–particle interaction contribution to F [Ch. 4, §4]; F_{id} ideal contribution to F [Ch. 4, §4)]; F_{M} magnetic contribution to F [Ch. 7, §5]; $F_{\mathrm{M}}^{\mathrm{at}}$ energy per atom due to magnetic ordering [Ch. 7, §5]
$\bar{F}(n,s)$	Laplace transform of $F(n,t)$, $\mathscr{L}(F(n,t))$
$F(n,t)$	$N(n,t)/N^{\mathrm{eq}}(n)$ [Ch. 2, §10; Ch. 3, §3; Ch. 3, §4]: $F^{\mathrm{st}}(n)$ steady-state solution of $F(n,t)$; $\Delta F^{\mathrm{st}}(n) = F^{\mathrm{st}}(n+1) - F^{\mathrm{st}}(n)$
g	Gibbs free energy per unit volume (free-energy density): g_0 for a system of uniform density [Ch. 4, §4]
$\mathfrak{g}$	surface coupling
g_{L}	Landé g-factor
G	Gibbs free energy (usually per mole)
$\mathfrak{G}$	applied strain-energy release rate [Ch. 12, §2]: $\mathfrak{G}_{\mathrm{emit}}$ critical rate for dislocation emission at a crack tip | temperature gradient at solidification front [Ch. 13, §2]

$G(n, \rho)$ correction for interfacial rates in coupled-flux nucleation that takes account of entropy change in the shell and original phase with the incorporation of an atom into the cluster interface

h enthalpy per unit volume | height of a spherical cap [Ch. 6, §2], normalized field, the functional analog to ln s [Ch. 10, §3] | Planck constant (6.626×10^{-34} J s) [Ch. 11, §6; Ch. 14, §3] | thickness of thin film [Ch. 12, §2]

$\mathfrak{h}$ surface field

$\hbar$ Planck constant divided by 2π

H enthalpy (usually per mole) | external applied magnetic field [Ch. 7, §5]: H_{appl} external applied; H_T total

$\mathscr{H}$ the Hamiltonian, $= \sum_i \dot{q}_i p_i - \mathscr{L}$ equal to the total energy

H_γ constant for the thickening rate of the γ product phase: $H_{\gamma\alpha}$ when affected by the atomic fluxes in the α phase

I nucleation rate or flux (usually $\text{mol}^{-1}\,\text{s}^{-1}$)

j flux of A atoms (shorthand for j^A)

$\mathbf{j}$ probability current; field flux

J total angular momentum [Ch. 7, Eq. (17)]

$J(n, t)$ flux of clusters past size n as a function of time [Ch. 2, §10]

k rate constant [Ch. 10, §7]: k_D for free diffusion on a lattice; k_{IA} for interfacial attachment | solute partition coefficient [Ch. 5, §4; Ch. 13, §2]: k^{eq} equilibrium value of coefficient [Ch. 5, §4]

k^+ forward rate constant

k^- backward rate constant

k_B Boltzmann constant ($1.38062 \times 10^{-23}\,\text{J K}^{-1}$)

K bulk modulus

$\bar{\mathbf{K}}$ tridiagonal rate-constant matrix

l^* a dimensionless edge-length of a cube-shaped critical cluster

ℓ thickness of lamella (polymer crystal)

L length of nucleus along a dislocation line [Ch. 6, §3] | orbital angular momentum [Ch. 7, §5] | edge length of the first Brillouin zone for the allowed sites in reciprocal space [Ch. 9, §2] | coherence length of lattice (polymer crystal) [Ch. 11, §6]

$\mathscr{L}$ the Lagrangian: kinetic energy minus potential energy

m mass: m, m_1 of a single molecule; m_A of a molecule of A; m_e of an electron | Avrami exponent [Ch. 8, §5; Ch. 9, §2] | liquidus slope (K at.%$^{-1}$ or K wt.%$^{-1}$) [Ch. 13, §2]

m_J projection of the total angular momentum J onto the direction of the external magnetic field H

M	order parameter: M_c value of M for which $\omega_l(M) = \omega_s(M)$ [Ch. 4, §4] | total magnetization [Ch. 7, §5] | number of independent generalized coordinates [Ch. 10, §2]
$\mathbf{M}$	generalized transport mobility matrix
M_s	martensite start temperature
n	number: of atoms or molecules in a cluster [Ch. 1, §2; Ch. 2, §2; Ch. 6, §4]; of methylene units in a polyethylene chain [Ch. 11, §6]; of planes in a stacking fault [Ch. 12, §4]; of molecules per unit volume [Ch. 14, §3]; of vacant lattice sites [Ch. 14, §5]
n^*	number of atoms or molecules in the critical nucleus
N	number (usually per mole): of clusters [Ch. 2, §2], of molecules [Ch. 4, §2], of droplets [Ch. 11, §2]; N_0 of single molecules in the original phase [Ch. 2, §2; Ch. 9, §2; Ch. 10, §3], of nucleant particles per unit volume [Ch. 13, §2]; N_1 of single molecules [Ch. 2, §4; Ch. 6, §1]; N_{amor} of still-uncrystallized (amorphous) droplets [Ch. 11, §2]; N_B of single molecules in contact with unit area of a boundary [Ch. 6, §2]; N_p of particles in Ostwald ripening [Ch. 4, §5]; N_S of excess surface atoms [Ch. 2, §2], of single molecules in contact with substrate per unit area of original phase [Ch. 6, §2]; N_{sol} of solute molecules per unit volume in the initial phase [Ch. 5, §5]; N_V of single molecules per unit volume [Ch. 6, §2; Ch. 9, §2], of atomic sites per unit volume [Ch. 14, §5]; N_{void} of voids per unit volume [Ch. 14, §5]
$\mathbb{N}$	time-dependent cluster population matrix
$N(n)$	cluster size distribution
N_A	Avogadro number (6.022142×10^{23} mol^{-1})
O	number of possible attachment sites: $O(a, b)$ on a cluster of a molecules of A and b molecules of B; $O(n)$ on a cluster of n atoms or molecules
O^*	total surface area of a critical cluster, $O(n^*)$
p	pressure
$\boldsymbol{p}$	vector momentum of a molecule
P	probability
$\mathscr{P}_{He}$	production rate of helium atoms under irradiation
q	single-molecule partition function [Ch. 4, §4] | scattering vector ($= 4\pi \sin\theta/\lambda$) [Ch. 7, §1]
q_i	generalized coordinates: $\dot{q}_i(= dq_i/dt)$ generalized velocities [Ch. 10, §2]
$\boldsymbol{q}_n$	reciprocal-lattice vectors
Q	canonical partition function [Ch. 4, §2; Ch. 7, §5; Ch. 10, §2] | strength of sources and sinks for solute diffusion [Ch. 4, §5] |

growth-restriction factor [Ch. 13, Eq. (2)] activation energy: Q_{rept} for reptational diffusion [Ch. 11, §6]; Q_0 for dislocation motion without applied stress [Ch. 12, §4]; Q_s for dislocation motion under applied stress [Ch. 12, §4]

r — radius of curvature of an interface; radius of a bubble, cluster, crystal, nucleus, particle, pore; distance from center of a cluster | ratio of interfacial attachment rates (k_B^+/k_A^+) [Ch. 5, §4]

$\boldsymbol{r}$ — position vector; vector from a site to its nearest-neighbor sites

r^* — critical radius for nucleation

R — universal gas constant (8.31434 J K^{-1} mol^{-1}) | lateral radius of/on a nucleant substrate: R_{Nuc} of planar particle face; R_p of a patch | R radius: of inscribing sphere [Ch. 6, Eq. (37)] of dislocation loop [Ch. 12, §2] | measured sample resistance [Ch. 9, §2]

$\boldsymbol{R}_i$ — vectors specifying real-space lattice sites

s — supersaturation | entropy per unit volume

$\mathfrak{s}$ — stress: $\mathfrak{s}_{res}$ resolved shear stress on glide plane; $\mathfrak{s}_\mu$ minimum, temperature-independent resistance to dislocation motion

s_i — spin state ($s_i = \pm 1$)

S — entropy (usually per mole): S_M^{at} entropy per atom due to magnetic ordering [Ch. 7, §5]; ΔS_M^{at} contribution of magnetic ordering to the difference in S_M^{at} between liquid and solid phases [Ch. 7, §5] | total spin angular momentum [Ch. 7, §5]

$S(a, b)$ — surface area of a cluster containing a molecules of A and b molecules of B [Ch. 5, §4]

$S(q)$ — X-ray structure factor as a function of scattering vector q [Ch. 4, §4]

t — time

T — temperature (typically in absolute units)

ΔT — supercooling ($= T_m - T$): ΔT_{max} maximum supercooling for solidification ($= T_m - T_{min}$) [Ch. 7, §2]; ΔT_N onset supercooling for nucleation [Ch. 13, §2]; ΔT_r reduced supercooling ($= \Delta T/T_m$ or $\Delta T/T_{liq}$) [Ch. 7, §2; Ch. 11, §2] | superheat ($= T_{max} - T_m$) [Ch. 14, §3]

T_g — glass-transition temperature: T_{rg} ($= T_g/T_m$) reduced glass-transition temperature

T_m — equilibrium melting temperature (typically in absolute units)

u — the ratio n/n^* [Ch. 3, §4] | cluster growth rate [Ch. 8, §5]

U — energy, internal energy of system | energy of a dislocation [Ch. 6, §3]: U_B a component of the strain energy; U_{core}

energy per unit length of the core | U_{es} electrostatic energy [Ch. 6, §4] | U_S energy per unit area by which metastable surface layer exceeds stable surface layer [Ch. 6, §2]

$\langle U \rangle$ average potential energy, [Ch. 10, Eq. (4)]

$\bar{v}$ atomic or molecular volume | partial molar volume [Ch. 5, §2]

v^* volume of critical cluster [Ch. 5, §2] | activation volume for dislocation nucleation [Ch. 12, §2]

$\mathbf{v}$ characteristic vectors: $\mathbf{v}_{cl}$ for the cluster; $\mathbf{v}_{liq}$ for the equilibrated liquid; $\mathbf{v}_{bcc}$ for the bcc phase; $\mathbf{v}_{ccp}$ for the ccp phase

V volume of sample/system

w number of available states

W work of cluster formation

W^* work of forming a critical cluster/nucleus (can also be represented as $W(n^*)$)

$W'(n)$ $W(n) - W(1)$ [Ch. 2, §2]

$W_{t,t'}(\xi \rightarrow \xi')$ transition probability [Ch. 4, §5]

$\delta W(n)$ the work of formation for a cluster of n+1 molecules less that of a cluster of n molecules

x volume fraction transformed [Ch. 8, §3] | number of gas atoms [Ch. 14, §5] | a distance coordinate [Ch. 15, §5]

X mole fraction | normalized radius of nucleant substrate ($= R_{Nuc}/r$) [Ch. 6, §2]

y distance from a dislocation line [Ch. 6, §3] | a spatial coordinate [Ch. 15, §4]

z height of pill-box embryo [Ch. 6, §2] | lattice coordination number [Ch. 9, §2]

z_A^* effective charge on A atoms within the product phase γ

Z Zeldovich factor [Ch. 2, Eq. (50)] [Ch. 2, §7; Ch. 9, §2; Ch. 14, §5] | partition function [Ch. 7, Eq. (17)] | constant in expression [Ch. 15, Eq. (26)] for the electromigration contribution to the intermixing flux of A and B in the product phase γ [Ch. 15, §5]

Z_N configurational integral [Ch. 4, §2]

$\alpha(n, \rho)$ rate at which solute atoms diffuse into the shell surrounding a cluster [Ch. 5, §5]

$\alpha(\phi)$ geometrical factor for the curved surface area of spherical-cap embryos [Ch. 6, §2]

α_{es} electrostatic energy factor [Ch. 6, Eq. (51)]

α_s supersaturation ratio for dissolved gas ($= C_g/C_0$) [Ch. 13, Eq. (12); Ch. 16, Eq. (3)]

β inverse thermal energy ($= (k_B T)^{-1}$) [Ch. 4, §2] | coefficient for the free-growth supercooling [Ch. 6, Eq. (37)] |