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Physics of Crystal Growth

This text introduces the physical principles of how and why crystals grow.

The first three chapters recall the fundamental properties of crystal surfaces at equilibrium. The next six chapters describe simple models and basic concepts of crystal growth including diffusion, thermal smoothing of a surface, and applications to semiconductors. Following chapters examine more complex topics such as kinetic roughness, growth instabilities, and elastic effects. A brief closing chapter looks back at the crucial contributions of crystal growth in electronics during this century. The book focuses on growth using molecular beam epitaxy. Throughout, the emphasis is on the role played by modern statistical physics. Informative appendices, interesting exercises and an extensive bibliography reinforce the text.

This book will be of interest to graduate students and researchers in statistical physics, materials science, surface physics and solid state physics. It will also be suitable for use as a coursebook at beginning graduate level.

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Physics of Crystal Growth

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Preface

In writing a preface, an author is faced with the question: what is this book of mine? Of course, in the end only the reader will decide what it really is. The scope of this preface, as of all prefaces, is to say what it was intended to be.

This book tries to offer a reasonably complete description of the physical phenomena which make solid materials grow in a certain way, homogeneous or not, rough or smooth. These phenomena belong to chemistry, quantum physics, mechanics, statistical mechanics. However, chemistry, mechanics and quantum physics are essentially the same during growth as they are at equilibrium. The statistical aspects are quite different. For this reason, the authors have insisted on statistical mechanics.

Another reason to emphasize the statistical mechanical concepts is that they will probably survive. The concepts developed many years ago by Frank, or more recently by Kardar, Parisi and Zhang are still valid while, for instance, quantum mechanical calculations of the relevant energy parameters will certainly evolve a lot in the next few years. We have not considered it useful to devote too many pages to them, but we have tried to present the frame in which the data can be inserted, as soon as they are known.

However, although emphasis is on statistical mechanics, other aspects are not ignored, even though they may have been treated somewhat superficially. The reader will find more detailed information in an extensive bibliography, where all titles are given in extenso, thus making its use much easier.

This book is mainly devoted to growth, and therefore to non-equilibrium processes. Nevertheless, we have tried to make it self-contained and to incorporate some elements of equilibrium surface physics, for instance the roughening transition and the equilibrium shape. The reader eager to know more will again find the necessary references in the bibliography.

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The authors are theorists and their book is mainly devoted to theory. Few details are given on experimental methods, but many experimental pictures (mostly from scanning tunneling microscopy) show how real materials do behave. In this domain, too, an abundant bibliography is available.

Although the responsability for all which is written here–good or bad– is completely ours, we owe a lot to all those who contributed to our understanding of the subject. We wish to thank J.M. Bermond, H. Bonzel, J.-P. Bucher, J. Chevrier, G. Comsa, J. Ernst, J. Frenken, M. Hanbücken, J.C. Heyraud, K. Kern, R. Kern, M. Lagally, J. Lapujoulade, J.J. Métois, B. Mutafschiev and E. Williams, whose experimental works revealed to us all the beauty of Surface Physics and Crystal Growth–and often contributed to the iconographic asset of the book. We are also very grateful to D. Wolf, Ph. Nozières, R. Kern again, J. Krug, P. Jensen, J. Langer, C. Misbah, L. Sander, D. Vvedensky and A. Zangwill, who shared with us some of their secrets. Special thanks are due to P. Politi and M. Schroeder, whose untiring reading of preliminary versions has been a source of most valuable suggestions and improvements. A good share of the chapters on elasticity has much profited from the competence of C. Duport, who corrected all our formulae, and even explained some of them to us!

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List of symbols

DLA: *Diffusion limited aggregation (section 11.2)*
MBE: *Molecular beam epitaxy*
ML: *Monolayer*
RHEED: *Reflection high-energy electron diffraction*

a: lattice constant or atomic distance (usually taken equal to 1 in this book).
A_α, $A_{\alpha\gamma}$, A: kinetic coefficients in sections 13.1 and 13.2
$A(t)$: amplitude appearing in section 13.7
B: a thermodynamic coefficient in (2.38)
$B_{\alpha\gamma}$: kinetic coefficient in chapter 13
C: a constant (section 15.7)
d: dimension of the space (usually 3)
d': surface dimension $d-1$
$d' = D/D'$, $d'' = D/D''$: see (6.20)
d_c^u: upper critical dimension (section 12.4)
d_c^ℓ: lower critical dimension (section 12.4)
d_f: fractal dimension of a fractal terrace (section 11.2)
$\tilde{D}$: Fick diffusion constant (sections 7.1, 8.4)
$\tilde{D}_s$, $\tilde{D}$: surface Fick diffusion constant
D^*: tracer diffusion constant (section 7.1)
$D_{\rm int}$: diffusion constant of interstitials
$D_{\rm vac}$: diffusion constant of vacancies in section 7.3.
D_s^0: see eq. (7.13)
D_0: see eq. (11.28)
D_2: diffusion constant of dimers at a surface (sections 7.10 and 11.8)
D', D'': step kinetic coefficients (section 6.4)
D_s (D when no ambiguity is possible): surface diffusion constant of adatoms
e *: basis of Napierian logarithms*
E: Young modulus
E_2: binding energy of a dimer (section 11.8)

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$\delta\mathbf{f}^{\text{ext}}$, $\delta\mathbf{F}^{\text{ext}}$: elementary external forces (chapter 16)
F_α^{ext}: External force
F: beam intensity in MBE
$\mathbf{F}_{\mathbf{R}}$: force acting at point $\mathbf{R}$
g: gravity (in chapter 1)
$G(\mathbf{R})$: height-height correlation function at equilibrium, eq. (1.4)
$G(\mathbf{r}, t)$ height-height correlation function during growth, eq. (12.1)
h: amplitude of the modulation of a surface, height of a defect
i^*: critical size of a cluster of adatoms (section 11.2)
$\mathbf{j}_s$: surface current density of adatoms
$\mathbf{j}_s^{\text{adat}}$: the current density of adatoms (section 7.5)
$\mathbf{j}_s^{\text{advac}}$: current density of advacancies (section 7.5)
$\mathbf{j}_k$: two-dimensional Fourier transform of the surface adatom current density
$\mathbf{k} = (k_x, k_y)$: a vector of the two-dimensional reciprocal space
k_B: the Boltzmann constant
K: bulk modulus (section 16.4)
K: kinetic coefficient defined by (12.6)
L_x, L_y: sample sizes in the x and y directions
L: linear size of a system
L: wavelength of a modulation (chapter 8)
$m_{\alpha\gamma}$, m: force dipole moment, eq. (15.2)
M: a point at the surface of a solid
$\mathbf{n}$: unit vector normal to a surface at a point, directed outward
N: number of particles in a physical system
N: quantity related to the size of the surface in section 2.6.
$N(t)$: cluster density (section 11.6)
$p_{\alpha\gamma}$, p_0: stress
$p_j(\mathbf{r}, t)$: probability that the j-th impurity is at $\mathbf{r}$ at time t (section 7.1)
$\mathbf{q} = (q_x, q_y)$: a vector of the two-dimensional reciprocal space
r_1: characteristic length (chapter 8)
R: radius of a terrace (section 8.5)
R: radius of curvature of a line or of a surface (chapter 10)
$\mathbf{R}$, $\mathbf{r}$: a point of the two-dimensional (chapter 1) or three-dimensional space
$\mathrm{d}S$: surface element (chapter 2)
S: surface of a crystal (chapters 3 and 4)
t: time
T: temperature
T_M: melting temperature.
T_R: roughening transition temperature
$u(\mathbf{r})$: atomic displacement at point $\mathbf{r}$
$\mathbf{v}$, $\mathbf{v}(\mathbf{n})$: velocity of the surface of a crystal (chapter 5)
v: velocity of a step
$v = V/N$: volume per atom (chapter 2)

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v_g: volume per atom in the vapour (chapter 2)
v_ℓ: volume per atom in the liquid (chapter 2).
V: volume
δV: volume element (section 16.1)
W: energy or free energy barrier
W_0: energy of a kink on a step (section 1.8)
W_0: energy barrier (section 15.7)
w_0: energy of a chemical bond (section 15.5)
W_a: adatom creation energy or free energy, see eq. (7.14a)
W_{adv}: see eq. (7.14b)
W_1: energy of a step per bond
W_{sd}: activation energy for surface diffusion (eq. (7.13) and section 11.8)
$W_{int}(r)$: interaction energy between defects at distance r (chapter 15)
W_{coh}: cohesive energy
x, y, z: coordinates of a point in the three-dimensional space.
$x_s = \sqrt{D_s \tau_v} = 1/\kappa$: average adatom diffusion length before desorption
y': derivative $\mathrm{d}y/\mathrm{d}x$
$\tilde{z}$: dynamical "critical exponent" for the correlation length (eq. 12.14)
$z_q(t)$; Fourier transform of $z(\mathbf{r}, t)$
j_s, D_s, Λ_s, $\tilde{D}_s$: section 8.4
α, γ: coordinates x, y or z
α: kinetic coefficient (eq. 7.8)
α: critical exponent for the spatial decay of the correlation function (eq. 12.18)
β: critical exponent for the temporal decay of the correlation function (eq. 12.18)
$\beta = 1/(k_B T)$
γ: free energy of a step per unit length or per atom (line tension)
$\tilde{\gamma} \equiv \gamma(\theta) + \mathrm{d}^2\gamma/\mathrm{d}\theta^2$: step stiffness (section 2.4)
γ: exponent defined in section 11.9
$\mathbf{\Gamma}(\mathbf{r} - \mathbf{r}')$: elastic Green function (eq. 15.10)
Γ: a step, in chapter 10
δa: misfit (section 15.4)
$\delta \mathscr{F}$, $\delta\mu$, etc.: increment of $\mathscr{F}$, μ, etc. from some reference value
$\delta\Sigma$: surface element (section 16.1)
ΔF: "supersaturation" $F - \rho_0/\tau_v$
Δ' and Δ'': kinetic coefficients defined in section 6.4
ϵ: Interaction energy between steps (section 1.7)
$\epsilon_{\alpha\gamma}(\mathbf{r})$: strain
$\epsilon(t)$: amplitude of a modulation in eq. (10.14) (called η in eq. (10.7))
ζ: Poisson ratio (eq. 6.12)
ζ: in section 10.6, a control parameter
η: amplitude of a modulation in eq. (10.7) (called ϵ in eq. (10.14))

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θ: value of an angle
$\kappa = 1/x_s$: inverse evaporation length of an adatom (eq. 6.6)
λ: capillary length (chapter 1)
λ: Lamé coefficient (chapters 15 and 6).
λ: kinetic coefficient in the KPZ equation (13.4)
λ: a constant (e.g. in section 4.2)
Λ_s, Λ: surface diffusion constant of advacancies (section 6.5)
μ: Lamé coefficient (chapters 15 and 16)
μ: chemical potential (elsewhere)
$\nu(t)$: number of sites visited by a diffusing adatom in time t (sections 6.3, 11.4)
ν: kinetic coefficient defined by (12.6)
$\xi(t)$: correlation length (section 12.6)
(Π): tangent plane to the crystal surface (chapter 3)
$\rho(\mathbf{r}, t)$, ρ_1, ρ_s: adatom density (occupied sites/surface sites)
ρ_n: surface density of clusters of n atoms (section 11.4).
ρ_0: equilibrium density of adatoms. (section 6.5)
$\bar{\rho}$: see section 6.3
$\rho_{\rm int}(\mathbf{r}, t)$: concentration of interstitials
σ_0: equilibrium density of advavancies (section 6.5)
σ, $\sigma(\mathbf{n})$: free energy of a surface per unit length or per atom (surface tension)
$\sigma(\mathbf{r})$: density of advacancies
Σ: Wulff's plot (section 3.2)
$\tilde{\sigma}$: surface rigidity, formula (2.5)
$1/\tau_v$: evaporation probability of an adatom per unit time (section 6.1)
$1/\tau_{\rm nuc}$: nucleation rate of new terraces (section 11.2)
$1/\tau_k$: the rate of emission of "gradatoms" (problem 10.6)
$\phi(z_x, z_y)$: the projected free energy density per unit area (eq. 2.2)
$\varphi\left(\{\epsilon(\mathbf{r})\}\right)$: elastic free energy density (section 16.5)
φ_p: components of an eigenvector of the transfer matrix in Appendix C
Φ: Gibbs free energy or free enthalpy $\mathscr{F} + PV$ (chapters 2 and 16)
ω: decay rate of a modulation, see eq. (10.15)
Ω: atomic area, generally set equal to 1 in this book
Ω: Grand potential $\mathscr{F} + PV - \mu N$
$\Omega^{\xi\zeta}_{\alpha\gamma}$: elastic constants
$\tilde{\Omega}^{\alpha\gamma}_{RR'}$: "discretized elastic constants", eq. (16.25)

ℓ: distance between steps
ℓ_s: adatom diffusion length before nucleation of a terrace (section 11.1)
ℓ_s: length on which diffusion is able to heal the surface (chapter 12)

$\mathrm{d}\mathscr{A}$: elementary projected area $\mathrm{d}x\mathrm{d}y$ (section 16.8)
$\mathscr{A}$: total projected area $L_x L_y$

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$\mathscr{F}$: *free energy*
$\mathscr{G}$: *Gibbs free energy* $\mathscr{G} = \mathscr{F} + PV$.
$\mathscr{N}$: *number of lattice sites on a surface in section 14.6*

$\simeq$: *nearly equal to (e.g.* $\sin x \simeq x$ *for small* x*)*
$\sim$: *proportional to (e.g.* $10x \sim x$*)*
$\approx$: *of the order of magnitude of (e.g.* $10 \approx 1$*)*

cot: *cotangent*
z_x, z_α: *partial derivative* $\partial z/\partial x$, $\partial z/\partial x_\alpha$
v_α, j_α, R_α: *the components of the vectors* $\mathbf{v}$, $\mathbf{j}$, $\mathbf{R}$
$\langle \chi \rangle$: *average value of a quantity* χ
$|z|$: *absolute value of* z
$\dot{z}$, $\dot{\rho}$, *etc.: derivative of* z, ρ, *etc., with respect to time*

$(hk\ell)$: *orientation of a crystal surface. E.g. the (001) face, the (111) orientation*
$[hk\ell]$: *orientation of a crystal axis. E.g.* $[110]$ *steps*
$\{hk\ell\}$: *set of crystal planes which are crystallographically equivalent. E.g. (cubic crystal): the* $\{001\}$ *orientations are the (001), (010) and (100) planes.*

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