Sculptured Thin Films
Nanoengineered Morphology and Optics
To

Mercedes N. Lakhtakia
Linda O. Messier

Oh, the THINKS
you can think up
if only you try!

Dr. Seuss (1904–1991)
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Preface

Everything is sculpture.
Any material, any idea without hindrance born into space,
I consider sculpture.

Isamu Noguchi (1904–1988)

Picking up a polished sample of ulexite, one of us thought that the parallel fibrous microstructure of this mineral was not unlike the matchstick morphology of columnar thin films shown to him by the other some years earlier. A telephone conversation led to a brainstorming session from which the mathematical concept of sculptured thin films—STFs, for short—began to emerge. That was in 1992. A 1966 paper of J.M. Niuwenhuizen and H.B. Haanstra provided the initial morphological bedrock, which was brought into optical focus by a 1989 paper of T. Motohiro and Y. Taga. Four years after our initial presentation in 1994 at a conference in France, and three years after our definitive presentation of the STF concept at Penn State, we stumbled upon a 1959 paper of N.O. Young and J. Kowal that provided an antecedental confirmation of the STF concept, which ties morphology and optics together.

Imagine a mass of isolated parallel matchsticks, all stuck on their lower ends to a glassy substrate. This arrangement describes columnar thin films. Imagine further that a still-life of the matchstick arrangement were painted by Salvador Dali: all the matchsticks—still isolated, still parallel to each other, and still propped upwards on the substrate—were depicted not straight but bent in some fanciful form. The imaginary Daliesque painting describes STFs. The matchsticks of cross-section diameter between 10 and 300 nm comprise clusters of sizes between 1 and 3 nm. Thus, the columns of an STF are shaped nanowires, and STFs can be considered as constituting a class of nanoengineered materials.

The optical response of a columnar thin film is like that of an orthorhombic crystal. Upon passage through a columnar thin film, the vibration ellipse of a plane wave is rotated and its axial ratio altered. Most importantly, a columnar thin film for optical purposes is effectively homogeneous. Think of a stack of extremely thin slices of an orthorhombic crystal. This stack is equivalent to a single-section STF for optical purposes. A single-section STF is therefore functionally nonhomoge-
neous in the thickness direction, but homogeneous in any transverse plane. In addition, it is effectively anisotropic. A multisection STF is a stack of single-section STFs. Upon passage through an STF, the vibration ellipse of a plane wave is rotated and its axial ratio altered in a desired fashion that can be incorporated by design into the morphology of the STF. There are, of course, nonoptical applications of STFs, which lie outside the scope of this book.

Our aim here is to provide the reader a basic knowledge of the morphology and the optical response characteristics of STFs, which are nanoeengineered by directional physical vapor deposition (PVD) onto substrates at oblique angles. While writing this book, our intent was not to simply compile and discuss the literature on STFs; in this day and age, that can be accomplished quite easily by anyone with the extensive electronic databases readily available. Rather, our intent was to lay the foundation for understanding thin film morphology so that scientists and technologists can design and engineer STF materials and devices for future applications, in particular optical applications. As such, the focus of this book is to couple the most detailed knowledge of thin film morphology—that includes the anisotropic, nanoscale clustering critical to STFs—with the response characteristics of optical STF devices.

We consciously avoided developments that we considered either too primitive or irrelevant to the theme of this book. For instance: (i) STFs made of magnetic materials have drawn some slight attention, but reported investigations are too rudimentary to be included in this text; (ii) photonic crystals made by directional PVD are not STFs due to inter-columnar spacing being at the wavelength scale; (iii) isotropic thin films are not STFs due to the absence of anisotropy; and (iv) organic STFs have not been included since only a couple of reports on polymeric STFs have been published to date. As research on STFs continues unabated, a complete treatment of all aspects of STFs is not possible at this time. We apologize if we left out a few topics dear to the reader’s heart, but let us also note that we left out some parts of our own recent research on STFs.

The STF concept has been resolutely mathematical from its emergence—to enable precise and predictable engineering of the vibration ellipse. Much experimental effort has been directed toward the realization of that goal. This we hope to have reflected in the 10 chapters of this book. Chapter 1 is a bird’s eye view of the past, the present, and the future of STFs, and is thus a book within a book. It can be read either all by itself or as an introduction to the following chapters. Chapters 2 to 5 focus on the shaped-nanowire morphology of STFs at the 1- to 1000-nm length scales, with emphasis on ways to achieve the desired morphology through simple movements of the substrate during growth. Chapters 6 to 10 focus on the optical properties of STFs, the effect of morphology on the reflection and transmission characteristics, and the principles underlying STF devices such as filters, polarizers, sensors, and radiators. Mathematica™ programs are provided in the text and on the accompanying CD so that the presented formalisms can be easily put to use. We expect that this book will enable the reader to select conditions to grow STFs with distinct morphologies, to understand opportunities and limitations of the
evolution of morphology, to solve electromagnetic equations in order to compute reflectances and transmittances, and eventually to engineering the morphology in order to fabricate optical STF devices with desirable polarization and bandwidth characteristics.

This book is aimed toward graduate students in optics at universities as well as toward practicing engineers in the optics industry. Expert researchers may find it useful in extending the STF concept and applications. Furthermore, we expect that the book is accessible to anyone who is interested in emerging nanotechnologies for optical devices as well as optics-based devices, provided he/she has taken typical undergraduate physics courses in optics and electromagnetism. Some knowledge of vectors, matrices, calculus, and differential equations is also necessary.

And now to the pleasant duty of acknowledging our debts of gratitude to many fine colleagues and friends:

Over the years, we have benefitted from collaborations with several leading researchers worldwide. For our work on STFs, we thank (in alphabetical order) Michael J. Brett (University of Alberta, Edmonton), Francesco Chiadini (Università di Salerno), Robert W. Collins (University of Toledo), Tariq Gilani (Millersville University), Ian J. Hodgkinson (University of Otago), Mark W. Horn (Penn State), Martin W. McCall (Imperial College London), John A. Polo, Jr. (Edinboro University of Pennsylvania), Kevin Robbie (Queen’s University, Kingston, Ontario), the late Werner S. Weiglhofer, and Qi hong Wu (University of Otago). Current and former students to whom we are indebted include Matthew D. Brubaker, Ryan J. Carey, Elif Ertekin, Craig Frankel, Joseph B. Geddes III, Thomas Gehrke, Ajay P. Giri, Robert Knepper, David P. Lewis, Mark W. Meredith, Jason T. Moyer, Steven F. Nagle, Frank Papa, Matthew D. Pickett, Wilfredo Otaño, Randy C. Ross, Pablo I. Rovira, Ronnen A. Roy, Joseph A. Sherwin, Erik E. Steltz, Paul D. Sunal, Philip Swab, Vijayakumar C. Venugopal, Bangyi Yang, Joseph E. Yehoda, Howard S. Witham, Fei Wang, and Jianwei Wang.

We are grateful to Álvaro Gómez (Universidad de Cantabria) and Mark W. Horn (Penn State) for supplying important illustrations, and to Alberto López Galindo (Universidad de Granada) for a sample of agate mineral. We thank Francesco Chiadini (Università di Salerno), Didier Felbacq (Université Montpellier II), Claes-Göran Granqvist (Uppsala Universitet), Tom G. Mackay (University of Edinburgh), and Walid Tabbara (Université Paris VI) for helping us locate old publications. We are indebted to Thomas Gehrke (Intrinsic Semiconductor) for translating old German publications.

Appreciation is extended to Craig F. Bohren (Penn State) for several discussions on planar optics during the course of writing this book, to S.V. Krishnaswamy (Northrop Grumman) for joint research on the morphology of thin films, and to Juan-Manuel García-Ruiz (Universidad de Granada) for a crucial collaboration on fractal morphology of materials. Also, we gratefully acknowledge our debt of gratitude to two prepublication reviewers.

We thank our colleagues, the late Werner S. Weiglhofer (University of Glasgow) and Francesco Costanzo (Penn State) for assistance in typesetting the book.
Thanks are due to Joseph B. Geddes III, Natalya S. Lakhtakia, Kelly Owens, Paul D. Sunal, Fei Wang, and Jian Xu for carefully going through various drafts of the manuscript. Penn State authorities kindly granted us both sabbatical leaves of absence for a semester, during which period a large part of this book was written. We also thank Richard P. McNitt and Judith A. Todd for sustained support of our STF research for many years.

Rick Hermann and Sharon Streams at SPIE supported our book proposal from its inception, while Margaret Thayer shepherded the production of the book, for which we are very grateful. We take this opportunity to also thank the office-bearers of SPIE for sustaining the scholarly endeavors of not only us but of many other colleagues worldwide.

Without the stability brought in our lives by our respective spouses, Mercedes Lakhtakia and Linda Messier, and their unstinted encouragement, this book would have taken several more years to write. To them this book is affectionately dedicated.

University Park, PA
October 2004

Akhlesh Lakhtakia
Russell Messier
## List of Acronyms

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<th>Description</th>
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<tr>
<td>AFM</td>
<td>atomic force microscope</td>
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<tr>
<td>CTF</td>
<td>columnar thin film</td>
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<tr>
<td>CVD</td>
<td>chemical vapor deposition</td>
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<tr>
<td>FIM</td>
<td>field ion microscope</td>
</tr>
<tr>
<td>HBM</td>
<td>helicoidal bianisotropic medium</td>
</tr>
<tr>
<td>ICM</td>
<td>isotropic chiral material</td>
</tr>
<tr>
<td>LC</td>
<td>liquid crystal</td>
</tr>
<tr>
<td>LCP</td>
<td>left circular polarization</td>
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<td>MODE</td>
<td>matrix ordinary differential equation</td>
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<tr>
<td>PSD</td>
<td>power spectral density</td>
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<tr>
<td>PVD</td>
<td>physical vapor deposition</td>
</tr>
<tr>
<td>RCP</td>
<td>right circular polarization</td>
</tr>
<tr>
<td>REM</td>
<td>replica electron microscopy</td>
</tr>
<tr>
<td>SAES</td>
<td>small-angle electron scattering</td>
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<tr>
<td>SEM</td>
<td>scanning electron microscope</td>
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<td>SNTF</td>
<td>sculptured nematic thin film</td>
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<tr>
<td>STF</td>
<td>sculptured thin film</td>
</tr>
<tr>
<td>SZM</td>
<td>structure zone model</td>
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<tr>
<td>TEM</td>
<td>transmission electron microscope</td>
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<tr>
<td>TFHBM</td>
<td>thin-film helicoidal bianisotropic medium</td>
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List of Principal Symbols and Operators

Symbols

0 null dyadic
[0] null matrix
\(a_{L,R}\) circular amplitudes of incident plane wave
\(a_{s,p}\) linear amplitudes of incident plane wave
\(\hat{a}_{mol}\) molar refractivity
\(A_{s,v}\) polarizability density dyadics
\([A]_{s,v}\) 6 \(\times\) 6 polarizability density matrixes
\(A_{L,R}\) circular absorbances
\(A_{s,p}\) linear absorbances
\(b_{L,R}\) circular amplitudes of emitted plane wave
\(\mathbf{b}, \mathbf{\hat{B}}\) microscopic, macroscopic magnetic fields
\(\mathbf{B}\) primitive magnetic field phasor
\([\mathbf{B}]\) 4 \(\times\) 4 rotation matrix
\(B_{L,R}\) emission efficiencies
\(c_{L,R}\) circular amplitudes of emitted plane wave
\(\hat{c}\) microscopic electric charge density
\([C]_{ref,s,v}\) 6 \(\times\) 6 constitutive matrixes
\(C_{L,R}\) emission efficiencies
\(\mathcal{C}D_{app}\) apparent circular dichroism
CDtru: true circular dichroism

D: induction electric field phasor

ds_{s-s}: distance between point source and substrate during deposition

d_{subs}: substrate diameter

\hat{D}: macroscopic induction electric field

\frac{D}{s,v}: depolarization dyadics

[D]_{s,v}: 6 \times 6 depolarization matrixes

e_{x,y,z}: Cartesian components of e

e, E: primitive electric field phasors

\tilde{e}, \tilde{E}: microscopic, macroscopic electric fields

f_v: void volume fraction; porosity

[f], [f'] : column vectors of size 4

[G], [G']: diagonal matrixes of eigenvalues

[\tilde{G}], [\tilde{G}']: spectral Green functions

h: structural handedness parameter

h_{x,y,z}: Cartesian components of h

h, H: induction magnetic field phasors

\hat{H}: macroscopic induction magnetic field

i = \sqrt{-1}

\frac{I}{}: identity dyadic

[I]: identity matrix

j_{so}, J_{so}: source electric current density phasors

\tilde{j}, \tilde{J}: microscopic, macroscopic electric current densities

\tilde{J}_{so}: macroscopic source electric current density

k_0: free-space wavenumber

[K]: 4 \times 4 matrix for plane waves

L: film thickness

L_{\Sigma}: thickness of a multilayer device
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<td>$\mathcal{LD}_{app}$</td>
<td>apparent linear dichroism</td>
</tr>
<tr>
<td>$\mathcal{LD}_{tru}$</td>
<td>true linear dichroism</td>
</tr>
<tr>
<td>$\tilde{M}$</td>
<td>macroscopic magnetization density</td>
</tr>
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<td>$[M], [M']$</td>
<td>$4 \times 4$ matrizes</td>
</tr>
<tr>
<td>$[M_{\Sigma}]$</td>
<td>$4 \times 4$ transfer matrix of a device</td>
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<tr>
<td>$n_{o,e}$</td>
<td>refractive indexes</td>
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<td>$\tilde{p}$</td>
<td>probability</td>
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<td>$p_{dep}$</td>
<td>pressure in deposition chamber</td>
</tr>
<tr>
<td>$\mathbf{p}, \mathbf{p}_0$</td>
<td>plane-wave polarization vectors</td>
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<td>$p_{vz}$</td>
<td>$z$-component of time-averaged Poynting vector of $\nu$th mode (chiral STF)</td>
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<td>$\tilde{\mathbf{P}}$</td>
<td>macroscopic polarization density</td>
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<td>$[P], [P']$</td>
<td>$4 \times 4$ matrix function</td>
</tr>
<tr>
<td>$\mathbf{r}$</td>
<td>position vector</td>
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<td>$r_{L,R}$</td>
<td>circular amplitudes of reflected plane wave</td>
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<td>$r_{LL,LR,RL,RR}$</td>
<td>circular reflection coefficients</td>
</tr>
<tr>
<td>$r_{s,p}$</td>
<td>linear amplitudes of reflected plane wave</td>
</tr>
<tr>
<td>$r_{ss,sp,ps,pp}$</td>
<td>linear reflection coefficients</td>
</tr>
<tr>
<td>$R_{LL,LR,RL,RR}$</td>
<td>circular reflectances</td>
</tr>
<tr>
<td>$R_{ss,sp,ps,pp}$</td>
<td>linear reflectances</td>
</tr>
<tr>
<td>$\mathbf{s}$</td>
<td>plane-wave polarization vector</td>
</tr>
<tr>
<td>$\tilde{\mathbf{S}}$</td>
<td>instantaneous Poynting vector</td>
</tr>
<tr>
<td>$\mathbf{S}$</td>
<td>rotation dyadic</td>
</tr>
<tr>
<td>$\mathbf{S}_{x,y,z}$</td>
<td>elementary rotation dyadics</td>
</tr>
<tr>
<td>$\hat{\mathbf{S}}_{y,z}$</td>
<td>rotation dyadics</td>
</tr>
<tr>
<td>$t$</td>
<td>time</td>
</tr>
<tr>
<td>$t_{L,R}$</td>
<td>circular amplitudes of transmitted plane wave</td>
</tr>
<tr>
<td>$t_{LL,LR,RL,RR}$</td>
<td>circular transmission coefficients</td>
</tr>
</tbody>
</table>
ts,p
linear amplitudes of transmitted plane wave

tss,sp,ps,pp
linear transmission coefficients

T
substrate temperature

T_{LL,LR,RL,RR}
circular transmittances

T_m
melting point

T_{ss,sp,ps,pp}
linear transmittances

T_{L,R}
circular total transmittances

T_{s,p}
linear total transmittances

u_{x,y,z}
Cartesian unit vectors

u_{\tau,n,b}
tangential, normal, and binormal unit vectors

U \equiv_{s,v}
ellipsoidal shape dyadics

[V], [V']
matrix containing eigenvectors

x, y, z
Cartesian coordinates

\alpha_{ref}
reference relative magnetoelectricity dyadic

\alpha_r
relative magnetoelectricity dyadic

\alpha_s
relative magnetoelectricity dyadic of deposited material

\alpha_v
relative magnetoelectricity dyadic in the void region

[\alpha]_{ref,s,v}
3 \times 3 matrix-equivalents of \alpha_{ref,s,v}

\beta_{ref}
reference relative magnetoelectricity dyadic

\beta_r
relative magnetoelectricity dyadic

\beta_s
relative magnetoelectricity dyadic of deposited material

\beta_v
relative magnetoelectricity dyadic in the void region

[\beta]_{ref,s,v}
3 \times 3 matrix-equivalents of \beta_{ref,s,v}

\gamma_{\tau,b}^{(s,v)}
ellipsoidal shape factors

\Gamma
gamma function
List of Principal Symbols and Operators

\(\delta_n\) linear birefringence

\(\delta_s\) ellipsoidal size measure

\(\delta_v\) significant extent of the \(\chi_v\)-distribution

\(\delta(\cdot)\) Dirac delta function

\(\Delta_{ba}\) anisotropy parameter

\((\Delta\lambda_0)^{Br}\) bandwidth of Bragg phenomenon (chiral STFs)

\(\epsilon_0\) permittivity of free space

\(\epsilon_{a,b,c}\) relative permittivity scalars

\(\epsilon_d\) composite relative permittivity scalar

\(\epsilon_r\) relative permittivity scalar (frequency domain)

\(\tilde{\epsilon}_r\) relative permittivity scalar (time domain)

\(\epsilon_{r}\) relative permittivity dyadic

\(\epsilon_{ref}\) reference relative permittivity dyadic

\(\epsilon_{ref}^o\) auxiliary relative permittivity dyadic

\(\epsilon_s\) relative permittivity dyadic of deposited material

\(\epsilon_v\) relative permittivity dyadic in the void region

\([[\epsilon]]_{ref,s,v}\) 3 \(\times\) 3 matrix-equivalents of \(\epsilon_{ref,s,v}\)

\(\zeta\) angular function

\(\eta_0\) intrinsic impedance of free space

\(\vartheta\) angle

\(\theta\) angle of incidence with respect to z axis

\(\kappa\) transverse wavenumber

\(\tilde{\kappa}\) cone-growth prefactor

\(\lambda_0\) free-space wavelength

\(\lambda_{0m}\) center-wavelength of Bragg phenomenon of order \(m\)

\(\lambda_0^{Br}\) center-wavelength Bragg phenomenon of order 2 (chiral STFs)

\(\mu_0\) permeability of free space
List of Principal Symbols and Operators

\( \mu_r \)  
relative permeability dyadic

\( \mu_{ref} \)  
reference relative permeability dyadic

\( \mu_s \)  
relative permittivity dyadic of deposited material

\( \mu_v \)  
relative permittivity dyadic in the void region

\([\mu]_{ref,s,v} \)  
3 × 3 matrix-equivalents of \( \mu_{ref,s,v} \)

\( \xi(z) \)  
angular function

\( \varphi \)  
angle

\( \rho_{so} \)  
source electric charge density phasor

\( \tilde{\rho} \)  
macroscopic electric charge density

\( \hat{\rho}_{mol} \)  
molar density

\( \tilde{\rho}_{so} \)  
macroscopic source electric charge density

\( \sigma \)  
dummy variable

\( \tilde{\sigma} \)  
variance of \( \varsigma \)

\( \varsigma \)  
cone-growth exponent

\( \bar{\varsigma} \)  
mean of \( \varsigma \)

\( \varsigma_d \)  
composite relative permittivity function

\( \tau \)  
angular function

\( \chi \)  
column inclination angle

\( \chi_v \)  
vapor incidence angle

\( \langle \chi_v \rangle \)  
average value of \( \chi_v \)

\( \chi_{vmb} \)  
maximum-bandwidth value of \( \chi_v \) (chiral STFs)

\( \chi_{vp} \)  
pseudoisotropic value of \( \chi_v \) (CTFs and chiral STFs)

\( \psi \)  
angle of incidence in \( xy \) plane

\( \omega \)  
angular frequency

\( \Omega \)  
structural period of C-shaped SNTF, structural half-period of chiral STF
Operators

\{\cdot\}^* \quad \text{complex conjugate}
\{\cdot\}^\dagger \quad \text{conjugate transpose}
\{\cdot\}^T \quad \text{transpose}
\{\cdot\}^{-1} \quad \text{inverse}
\text{Im}\{\cdot\} \quad \text{imaginary part of}
\mathcal{P} \quad \text{principal value}
\text{Re}\{\cdot\} \quad \text{real part of}