Emerging Trends in Research on Sculptured Thin Films

Akhlesh Lakhtakia

Department of Engineering Science and Mechanics
The Pennsylvania State University

May 26, 2006
Instituto de Investigaciones en Materiales
Universidad Nacional Autónoma de México
Ciudad de México
Thanks

- Carlos I. Mendoza
- IIM, UNAM
- J. Adrian Reyes Cervantes
- IF, UNAM
Collaborators

- Mark W. Horn (Penn State)
- Jian Xu (Penn State)
- Melik C. Demirel (Penn State)
- J. Adrian Reyes (IF, UNAM)
Outline

• Introduction
• Optical Applications
• Optical Modeling
• Emerging Directions
  – Light Emitters
  – STFs with Gain
  – Electrically Controlled STFs
  – Polymeric STFs
  – Bioscaffolds
  – STFs with Transverse Architecture
INTRODUCTION
Sculptured Thin Films

Assemblies of Parallel Curved Nanowires/Submicronwires

Controllable Nanowire Shape
Sculptured Thin Films

Assemblies of Parallel Curved Nanowires/Submicronwires

Controllable Nanowire Shape

2-D - nematic
3-D - helicoidal
Sculptured Thin Films

Assemblies of Parallel Curved Nanowires/Submicronwires

Controllable Nanowire Shape

2-D - nematic
3-D - helicoidal
combination morphologies
vertical sectioning
Sculptured Thin Films

Assemblies of Parallel Curved Nanowires/Submicronwires

Controllable Nanowire Shape

2-D - nematic
3-D - helicoidal

combination morphologies
vertical sectioning

Nanoengineered Materials (1-3 nm clusters)

Controllable Porosity (10-90 %)
Sculptured Thin Films

Antecedents:

(i) Young and Kowal - 1959

(ii) Niuwenhuizen & Haanstra - 1966

(iii) Motohiro & Taga - 1989

Conceived as an optical material by Lakhtakia & Messier (1992-1995)
Sculptured Thin Films

Collaborators:

(i) Weiglhofer, University of Glasgow

(ii) Robbie & Brett, University of Alberta

(iii) McCall, Imperial College London

(iv) Hodgkinson, University of Otago

(v) Penn State Colleagues & Students
Physical Vapor Deposition
(Columnar Thin Films)
Physical Vapor Deposition
(Sculptured Thin Films)

- Rotate about y axis for nematic morphology
- Rotate about z axis for helicoidal morphology
- Mix and match rotations for complex morphologies
Physical Vapor Deposition
(Serial Bideposition)

Adapted for STFs by Hodgkinson
## Sculptured Thin Films

### Optical Devices:
- Polarization Filters
- Bragg Filters
- Ultranarrowband Filters
- Fluid Concentration Sensors
- Bacterial Sensors

### Biomedical Applications:
- Tissue Scaffolds
- Drug/Gene Delivery
- Bone Repair
- Virus Traps

### Other Applications
OPTICAL APPLICATIONS
Chiral STFs: Circular Bragg Phenomenon

A simple explanation (Coupled-Wave Theory):

- Co-handed wave: Scalar Bragg grating
- Cross-handed wave: Homogeneous bulk medium
Chiral STF as CP Filter

Figure 10.2: Predicted and measured transmittances of a circular polarization filter as functions of the free-space wavelength $\lambda_0$ for normal incidence. The filter is a chiral STF of patinal titanium oxide. The reference permittivity dyadic was predicted with $\varepsilon_r = 6.3 + i0.012$, $\varepsilon_v = 1$, $f_v = 0.421$, $\gamma^{(a)} = \gamma^{(b)} = 20$, and $\gamma^{(a)} = \gamma^{(b)} = 1.06$ set in Program 6.1. The other parameters are $\chi = 47$ deg, $h = -1$, $\Omega = 173$ nm, $L = 30$ $\Omega$, and $\psi = 0$ deg. (Adapted from Sherwin et al. [109] with permission of Elsevier.)
Spectral Hole Filter

Figure 10.10: Measured transmittances of a narrow bandpass filter comprising an isotropic homogeneous spacer of hafnium oxide interposed between two identical, structurally left-handed, chiral STF sections of titanium oxide. Evidence of a hole in the spectrum of $R_{LL}$ at 580-nm wavelength is provided by the spectrum of $T_{LL}$. (Adapted from Hodgkinson et al. [125] with permission of Elsevier.)
Fluid Concentration Sensor

Figure 10.22: Optical response of a narrow bandpass filter, described by Eq. (10.17) and made of two structurally left-handed chiral STF sections, on infiltration by water vapor. The dotted lines indicate the measured transmittance spectrum when the filter was dry. The filter was flooded with water and then allowed to recover by evaporation in air. Transmittance spectrums recorded at 5-s intervals after the flooding are shown. (Adapted from Lakhtakia et al. [105] with permission of Elsevier.)
OPTICAL MODELING
Optical Modeling of STFs

\[
D(r, \omega) = \varepsilon_0 S(z) \cdot \left[ \varepsilon_{\text{ref}}(\omega) \cdot S^T(z) \cdot E(r, \omega) + \alpha_{\text{ref}}(\omega) \cdot S^T(z) \cdot H(r, \omega) \right],
\]

\[
B(r, \omega) = \mu_0 S(z) \cdot \left[ \beta_{\text{ref}}(\omega) \cdot S^T(z) \cdot E(r, \omega) + \mu_{\text{ref}}(\omega) \cdot S^T(z) \cdot H(r, \omega) \right],
\]

\[
S_x(z) = u_x u_x + (u_y u_y + u_z u_z) \cos \xi(z) + (u_z u_y - u_y u_z) \sin \xi(z),
\]

\[
S_y(z) = u_y u_y + (u_x u_x + u_z u_z) \cos \tau(z) + (u_z u_x - u_x u_z) \sin \tau(z),
\]

\[
S_z(z) = u_z u_z + (u_x u_x + u_y u_y) \cos \zeta(z) + (u_y u_x - u_x u_y) \sin \zeta(z).
\]
Optical Modeling of STFs

Dielectric Materials

\[ D(r, \omega) = \varepsilon_0 \varepsilon_r (z, \omega) \cdot E(r, \omega) \]
\[ = \varepsilon_0 S(z) \cdot \varepsilon_{ref}(\omega) \cdot S^T(z) \cdot E(r, \omega), \]
\[ B(r, \omega) = \mu_0 H(r, \omega). \]
Optical Modeling of STFs

Locally Orthorhombic Materials

\[ D(r, \omega) = \varepsilon_0 \varepsilon_r(z, \omega) \cdot E(r, \omega) \]
\[ = \varepsilon_0 S(z) \cdot \varepsilon_{ref}(\omega) \cdot S^T(z) \cdot E(r, \omega), \]
\[ B(r, \omega) = \mu_0 H(r, \omega). \]

\[ \varepsilon_{ref}(\omega) = \hat{S}_y(\chi) \cdot \varepsilon^{o}_{ref}(\omega) \cdot \hat{S}^T_y(\chi) \]

\[ \varepsilon^{o}_{ref}(\omega) = \varepsilon_{ref}(\omega) \bigg|_{\chi=0} = \varepsilon_a(\omega) u_z u_z + \varepsilon_b(\omega) u_x u_x + \varepsilon_c(\omega) u_y u_y \]

\[ \hat{S}_y(\chi) = u_y u_y + (u_x u_x + u_z u_z) \cos \chi + (u_z u_x - u_x u_z) \sin \chi \]
Optical Modeling of STFs

Homogenize a collection of parallel ellipsoids to get

\[ \varepsilon_{ref}(\omega) \]

Optical Modeling of STFs

Wave Propagation

\[ E(\mathbf{r}, \omega) = e(z, \kappa, \psi, \omega) \exp[i\kappa(x \cos \psi + y \sin \psi)] \]

\[ H(\mathbf{r}, \omega) = h(z, \kappa, \psi, \omega) \exp[i\kappa(x \cos \psi + y \sin \psi)] \]

\[ \nabla \times E(\mathbf{r}, \omega) = i\omega B(\mathbf{r}, \omega), \]

\[ \nabla \times H(\mathbf{r}, \omega) = -i\omega D(\mathbf{r}, \omega), \]

\[ \frac{d}{dz} \mathbf{f}(z, \kappa, \psi, \omega) = i\mathbf{P}(z, \kappa, \psi, \omega) \mathbf{f}(z, \kappa, \psi, \omega). \]

\[ \mathbf{f}(z, \kappa, \psi, \omega) = \begin{bmatrix} e_x(z, \kappa, \psi, \omega) \\ e_y(z, \kappa, \psi, \omega) \\ h_x(z, \kappa, \psi, \omega) \\ h_y(z, \kappa, \psi, \omega) \end{bmatrix} \]
EMERGING DIRECTIONS
1. LIGHT EMITTERS
LIGHT EMITTERS

- Luminophores inserted in a chiral STF
- Co- and contra-wound photonic source filaments
- Calculations using Maxwell postulates
  - volume fraction of filaments
  - wavelength
  - co/contra-wound
Fig. 1. Computed spectrums of the emission efficiencies $\beta_{R,L}$ and $\gamma_{R,L}$ as functions of the fraction $f$ of a chiral STF occupied by co-wound photon source filaments and the free-space wavelength $\lambda_0$. See the text for the constitutive and other parameters used. The Bragg regime for the selected parameters is $\lambda_0 \in [513.4, 531.8]$ nm.
LIGHT EMITTERS

Contra-wound

Fig. 2. Same as Fig. 1, except that the photon source filaments are contra-wound.
LIGHT EMITTERS

• Co/contra-wound:

  Clear differences in
  (i) polarization state
  (ii) emission bandwidth

• Dependence on tilt angle $\chi$

   - nonlinear
LIGHT EMITTERS

• Luminophores (Alq3) inserted in a cavity between two chiral STFs
LIGHT EMITTERS

- Luminophores (Alq3) inserted in a cavity between two chiral STFs
LIGHT EMITTERS

- Luminophores (Alq3) inserted in a cavity between two chiral STFs

Fig. 4. Schematic of the optical bench used to characterize the circular polarization light-emitting device. The inset shows how a quarter-wave Fresnel-rhomb retarder converts LCP and RCP light beams to the linearly polarized light beams whose polarization directions are orthonormal.
LIGHT EMITTERS

- Luminophores (Alq3) inserted in a cavity between two left-handed chiral STFs
2. STFs WITH GAIN
STFs WITH GAIN

- Chiral STF

\[ \varepsilon^0_{\text{ref}}(\omega) = \varepsilon_{\text{ref}}(\omega) \bigg|_{\chi=0} = \varepsilon_a(\omega) \mathbf{u}_z \mathbf{u}_z + \varepsilon_b(\omega) \mathbf{u}_x \mathbf{u}_x + \varepsilon_c(\omega) \mathbf{u}_y \mathbf{u}_y \]

- Solve Maxwell postulates for reflection and transmission

\[ \varepsilon_a = 2.5 (1 + i\delta_\varepsilon), \quad \varepsilon_b = 3.2 (1 + i\delta_\varepsilon), \quad \varepsilon_c = 2.6 (1 + i\delta_\varepsilon) \]

- \( \delta_\varepsilon > 0 \) absorption
- \( \delta_\varepsilon < 0 \) gain
- \( \delta_\varepsilon = 0 \) no absorption, no gain
STFs WITH GAIN

No loss, no gain

Loss

Gain

Lakhtakia & Xu, at press
STFs WITH GAIN

No loss, no gain

Gain

High density of states

Normalized Wavelength $\lambda_0 / \Omega$
STFs WITH GAIN

High Density of States

implies

High Emission

Analogy: Lasing by dye-doped CLCs
3. ELECTRICALLY CONTROLLED STFs
ELECTRICALLY CONTROLLED STFs

\[
\begin{pmatrix}
\frac{1}{\varepsilon_1^{(0)}} + \sum_{K=1}^{3} r_{1K} E_{K}^{dc} & \sum_{K=1}^{3} r_{6K} E_{K}^{dc} & \sum_{K=1}^{3} r_{5K} E_{K}^{dc} \\
\sum_{K=1}^{3} r_{6K} E_{K}^{dc} & \frac{1}{\varepsilon_2^{(0)}} + \sum_{K=1}^{3} r_{2K} E_{K}^{dc} & \sum_{K=1}^{3} r_{4K} E_{K}^{dc} \\
\sum_{K=1}^{3} r_{5K} E_{K}^{dc} & \sum_{K=1}^{3} r_{4K} E_{K}^{dc} & \frac{1}{\varepsilon_3^{(0)}} + \sum_{K=1}^{3} r_{3K} E_{K}^{dc}
\end{pmatrix}
\]

\[
\frac{\varepsilon^{0}}{\varepsilon_{r e f}} = \frac{\varepsilon}{\varepsilon_{P E}}
\]

DC voltage across the thickness
Electrically controlled optical bandgap in a structurally chiral material

J. Adrian Reyes a,b, Akhlesh Lakhtakia b,c,*

a Instituto de Física, Universidad Nacional Autónoma de México, Aparato Postal 20-364, C.P. 01000, Mexico D.F., Mexico
b Computational and Theoretical Materials Sciences Group (CATMAS), Department of Engineering Science and Mechanics, Pennsylvania State University, 212 EES Building, University Park, PA 16802-6812, USA
c Photonics Section, Department of Physics, Imperial College, London SW7 2AZ, United Kingdom

Also:
(2) Lakhtakia & Reyes, Phys. Rev. E, submitted
ELECTRICALLY CONTROLLED STFs

Without dc voltage

With dc voltage
ELECTRICALLY CONTROLLED STFs

Without dc voltage

Pseudo-Isotropic point
ELECTRICALLY CONTROLLED STFs

Without dc voltage

With dc voltage

Pseudo-Isotropic point
ELECTRICALLY CONTROLLED STFs

- Pseudo-Isotropic point

Without dc voltage

With dc voltage
4. POLYMERIC STFs
POLYMERIC STFs

1. Replamineform (Multi-Step) Technique
2. Combined CVD-PVD Technique
3. Holographic Lithography
POLYMERIC STFs: REPLAMINEFORM TECHNIQUE

Suggestion published in 1996

© World Scientific Publishing

SCULPTURED THIN FILMS (STFS) FOR OPTICAL, CHEMICAL AND BIOLOGICAL APPLICATIONS

A. Lakhtakia,¹ R. Messier,¹,² M. J. Brett³ and K. Robbie³
POLYMERIC STFs: REPLAMINEFORM TECHNIQUE (3-step)

Implementation published in 2001:


Three-step procedure:

1. Make a chiral STF
2. Fill the void regions with a polymer
3. Etch out the skeleton material

Helical holes

Elias, Harris, Brett, *J.M.S.* 13, 808 (2004)
POLYMERIC STFs: REPLAMINEFORM TECHNIQUE (3-step)

Helical holes

Excellent for piezoelectrically controlled STFs

Elias, Harris, Brett, *J.M.S.* 13, 808 (2004)

On piezoelectric control of the optical response of sculptured thin films

FEI WANG, AKHLESH LAKHTAKIA¹ and RUSSELL MESSIER
POLYMERIC STFs:
REPLAMINEFORM TECHNIQUE (5-step)

5-step procedure:

1. Make a chiral STF
2. Fill the void regions with polymer A
3. Etch out the skeleton material
4. Fill the void region regions with polymer B
5. Etch out polymer A
POLYMERIC STFs: COMBINED CVD+PVD TECHNIQUE

1-Step Process

First, pyrolize to monomer state, and then deposit
POLYMERIC STFs: COMBINED CVD+PVD TECHNIQUE

The PDS 2010 system and the schematic of stepper motor and nozzle assembly used with the polymer deposition system.
1. 4-laser beams to expose photoresist
   (1 beam should be elliptically polarized)

2. Develop the exposed photoresist layer
POLYMERIC STFs:
HOLOGRAPHIC LITHOGRAPHY

Photonic Crystals vs. STFs
5. BIOSCAFFOLDS
The three advantages of STFs are as follows:

1. **Surface-to-volume ratio is very high in STF films (> two orders of magnitude).**

2. **STFs can be made out of virtually any material and can be endowed with transverse architectures to provide the best possible substrates for attachment at the nanoscale.**

3. **Optical properties suitable for sensing.**

BIOSCAFFOLDS

Cells grow, but detach

Cells grow, and attach
BIOSCAFFOLDS

Cell growth

Red: Nucleus

Green: F-actin

Human 293-Kidney Cells

Z Position z (1 µm)
BIOSCAFFOLDS

![Graph showing cell area coverage over time for HEK-293 cell growth without serum. The graph displays bars for time points 48, 72, and 96 hours.]
6. STFs WITH TRANSVERSE ARCHITECTURE
STFs WITH TRANSVERSE ARCHITECTURE

STFs on Microscale Topography
(Cross-sectional SEMs of SiOx STFs)
STFs WITH TRANSVERSE ARCHITECTURE

Metal STFs on Topography

Chromium

Aluminum

Molybdenum
STFs WITH TRANSVERSE ARCHITECTURE

Semiconductor STFs on Micro and Nanoscale Topography

SnO$_x$ STFs grown on photoresist patterns
Sculptured Nanowires on Nanoscale Topography

- Single SiOx nanowire array grown on 60 nm e-beam resist
- HCP array of SiOx nanocolumns
- BCC array of SiOx nanocolumns
- 1um x 1um mesh of SiOx nanolines
STFs WITH TRANSVERSE ARCHITECTURE

Blending of nanoscale and microscale in uniform large-area sculptured thin-film architectures

Mark W Horn, Matthew D Pickett, Russell Messier and Akhlesh Lakhtakia

Department of Engineering Science and Mechanics, Pennsylvania State University, University Park, Pennsylvania 16802

(Received 25 June 2004; accepted 4 October 2004; published 14 December 2004)

We have grown helicoidal nanowire assemblies on a variety of topographic substrates with regular microlithographic patterns, thereby demonstrating that sculptured thin films with transversely latticed architecture can be grown by physical vapor deposition. The transverse feature-separations are as low as 100–300 nm, and mesa regions are circular posts as small as 60 nm in diameter. The initial as well as the subsequent stages of growth on topographic substrates can be understood using simple geometric shadowing arguments. © 2004 American Vacuum Society.
Emerging Directions

- Light Emitters
- STFs with Gain
- Electrically Controlled STFs
- Polymeric STFs
- Bioscaffolds
- STFs with Transverse Architecture
Muchas gracias