

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 Ciuded 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

Sal 16

Sculptured Thin Films

Nanoengineered Morphology and Optics



SPIE Press (2005)

Assemblies of Parallel Curved Nanowires/Submicronwires

Controllable Nanowire Shape





Assemblies of Parallel Curved Nanowires/Submicronwires

Controllable Nanowire Shape

2-D - nematic3-D - helicoidal





Assemblies of Parallel Curved Nanowires/Submicronwires

Controllable Nanowire Shape

 2-D - nematic
 3-D - helicoidal combination morphologies
 vertical sectioning

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 %)

Antecedents:

- (i) Young and Kowal 1959
- (ii) Niuewenhuizen & Haanstra 1966
- (iii) Motohiro & Taga 1989

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

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)





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 λ_0 for normal incidence. The filter is a chiral STF of patinal titanium oxide. The reference permittivity dyadic was predicted with $\epsilon_s = 6.3 + i0.012$, $\epsilon_v = 1$, $f_v = 0.421$, $\gamma_{\tau}^{(s)} = \gamma_{\tau}^{(v)} = 20$, and $\gamma_b^{(s)} = \gamma_b^{(v)} = 1.06$ set in Program 6.1. The other parameters are $\chi = 47 \text{ deg}$, h = -1, $\Omega = 173 \text{ nm}$, $L = 30 \Omega$, and $\psi = 0 \text{ deg}$. (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

$$\begin{split} \mathbf{D}(\mathbf{r},\omega) &= \epsilon_0 \,\underline{\underline{S}}(z) \bullet \left[\underline{\underline{\epsilon}}_{ref}(\omega) \bullet \underline{\underline{S}}^T(z) \bullet \mathbf{E}(\mathbf{r},\omega) \right. \\ &\quad + \underline{\underline{\alpha}}_{ref}(\omega) \bullet \underline{\underline{S}}^T(z) \bullet \mathbf{H}(\mathbf{r},\omega) \right], \\ \mathbf{B}(\mathbf{r},\omega) &= \mu_0 \,\underline{\underline{S}}(z) \bullet \left[\underline{\underline{\beta}}_{ref}(\omega) \bullet \underline{\underline{S}}^T(z) \bullet \mathbf{E}(\mathbf{r},\omega) \right. \\ &\quad + \underline{\underline{\mu}}_{ref}(\omega) \bullet \underline{\underline{S}}^T(z) \bullet \mathbf{H}(\mathbf{r},\omega) \right], \end{split}$$

Linear Bianisotropic Materials



Introduction to Complex Mediums for Optics and Electromagnetics

Editors: Werner S. Weiglhofer • Akhlesh Lakhtakia



$$\begin{split} \underline{\underline{S}}_{x}(z) &= \mathbf{u}_{x}\mathbf{u}_{x} + (\mathbf{u}_{y}\mathbf{u}_{y} + \mathbf{u}_{z}\mathbf{u}_{z})\,\cos\,\xi(z) \\ &+ (\mathbf{u}_{z}\mathbf{u}_{y} - \mathbf{u}_{y}\mathbf{u}_{z})\,\sin\,\xi(z)\,,\\ \underline{\underline{S}}_{y}(z) &= \mathbf{u}_{y}\mathbf{u}_{y} + (\mathbf{u}_{x}\mathbf{u}_{x} + \mathbf{u}_{z}\mathbf{u}_{z})\,\cos\,\tau(z) \\ &+ (\mathbf{u}_{z}\mathbf{u}_{x} - \mathbf{u}_{x}\mathbf{u}_{z})\,\sin\,\tau(z)\,,\\ \underline{\underline{S}}_{z}(z) &= \mathbf{u}_{z}\mathbf{u}_{z} + (\mathbf{u}_{x}\mathbf{u}_{x} + \mathbf{u}_{y}\mathbf{u}_{y})\,\cos\,\zeta(z) \\ &+ (\mathbf{u}_{y}\mathbf{u}_{x} - \mathbf{u}_{x}\mathbf{u}_{y})\,\sin\,\zeta(z)\,. \end{split}$$

SPIE Press (2003)

Dielectric Materials

$$\begin{aligned} \mathbf{D}(\mathbf{r},\omega) &= \epsilon_0 \, \underline{\epsilon}_r \, (z,\omega) \, \bullet \, \mathbf{E}(\mathbf{r},\omega) \\ &= \epsilon_0 \, \underline{\underline{S}}(z) \, \bullet \, \underline{\underline{S}}_{ref}(\omega) \, \bullet \, \underline{\underline{S}}^T(z) \, \bullet \, \mathbf{E}(\mathbf{r},\omega) \,, \\ \mathbf{B}(\mathbf{r},\omega) &= \mu_0 \, \mathbf{H}(\mathbf{r},\omega) \,. \end{aligned}$$

Locally Orthorhombic Materials

$$\begin{aligned} \mathbf{D}(\mathbf{r},\omega) &= \epsilon_0 \,\underline{\epsilon}_r \left(z, \omega \right) \, \bullet \, \mathbf{E}(\mathbf{r},\omega) \\ &= \epsilon_0 \,\underline{\underline{S}}(z) \, \bullet \,\underline{\underline{S}}^T(z) \, \bullet \, \mathbf{E}(\mathbf{r},\omega) \,, \\ \mathbf{B}(\mathbf{r},\omega) &= \mu_0 \, \mathbf{H}(\mathbf{r},\omega) \,. \end{aligned}$$

$$\underline{\underline{\epsilon}}_{ref}(\omega) = \underline{\underline{\hat{S}}}_{y}(\chi) \bullet \underline{\underline{\hat{S}}}_{ref}^{o}(\omega) \bullet \underline{\underline{\hat{S}}}_{y}^{T}(\chi)$$

$$\underbrace{\epsilon_{ref}^{o}}_{e_{ref}}(\omega) = \underbrace{\epsilon_{ref}}_{e_{ref}}(\omega) \Big|_{\chi=0} = \epsilon_{a}(\omega) \mathbf{u}_{z} \mathbf{u}_{z} + \epsilon_{b}(\omega) \mathbf{u}_{x} \mathbf{u}_{x} + \epsilon_{c}(\omega) \mathbf{u}_{y} \mathbf{u}_{y}$$

$$\underline{\hat{S}}_{y}(\chi) = \mathbf{u}_{y}\mathbf{u}_{y} + (\mathbf{u}_{x}\mathbf{u}_{x} + \mathbf{u}_{z}\mathbf{u}_{z})\cos\chi + (\mathbf{u}_{z}\mathbf{u}_{x} - \mathbf{u}_{x}\mathbf{u}_{z})\sin\chi$$



Homogenize a collection of parallel ellipsoids to get $\underline{\epsilon}_{ref}^{o}(\omega)$



Sherwin and Lakhtakia (2001-2003): Bruggeman formalism

Mathematica Program

Optical Modeling of STFs Wave Propagation

 $\begin{aligned} \mathbf{E}(\mathbf{r},\omega) &= \mathbf{e}(z,\kappa,\psi,\omega) \, \exp\left[i\kappa(x\cos\psi+y\sin\psi)\right] \\ \mathbf{H}(\mathbf{r},\omega) &= \mathbf{h}(z,\kappa,\psi,\omega) \, \exp\left[i\kappa(x\cos\psi+y\sin\psi)\right] \end{aligned}$

$$\nabla \times \mathbf{E}(\mathbf{r}, \omega) = i\omega \mathbf{B}(\mathbf{r}, \omega),$$
$$\nabla \times \mathbf{H}(\mathbf{r}, \omega) = -i\omega \mathbf{D}(\mathbf{r}, \omega),$$



Sculptured Thin Films



Nanoengineered Morphology and Optics

Akhlesh Lakhtakia Russell Messier

> Mathematica Program

$$[\mathbf{f}(z,\kappa,\psi,\omega)] = \begin{bmatrix} e_x(z,\kappa,\psi,\omega) \\ e_y(z,\kappa,\psi,\omega) \\ h_x(z,\kappa,\psi,\omega) \\ h_y(z,\kappa,\psi,\omega) \end{bmatrix}$$

EMERGING DIRECTIONS

- 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

Co-wound



Fig. 1. Computed spectrums of the emission efficiencies $\mathscr{B}_{R,L}$ and $\mathscr{C}_{R,L}$ as functions of the fraction f of a chiral STF occupied by cowound photon source filaments and the free-space wavelength λ_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.

Contra-wound



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

• Co/contra-wound:

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

• Dependence on tilt angle χ

- 1. Lakhtakia, Opt. Commun. 188, 313 (2001)
- 2. Lakhtakia, Opt. Commun. 202, 103 (2002)
- 3. Lakhtakia, MOTL 37, 37 (2003)
- 4. Steltz & Lakhtakia, *Opt. Commun.* **216**, 139 (2003) nonlinear
Luminophores (Alq3) inserted in a cavity between two chiral STFs

OPTICS 11240 2 March 2006 Disk Used	ARTICLE IN PRESS	No. of Pages 5, Model 5+
	Available online at www.sciencedirect.com	Optics Communications
ELSEVIER	Optics Communications xxx (2006) xxx-xxx	www.elsevier.com/locate/optcom

Circularly polarized fluorescence from light-emitting microcavities with sculptured-thin-film chiral reflectors

Jian Xu^{a,*}, Akhlesh Lakhtakia^a, Justin Liou^a, An Chen^a, Ian J. Hodgkinson^b

^a Department of Engineering Science and Mechanics, Pennsylvania State University, University Park, PA 16802-6812, USA ^b Department of Physics, University of Otago, P.O. Box 56, Dunedin, New Zealand

Received 26 October 2005; received in revised form 4 January 2006; accepted 9 February 2006

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



Fig. 3. Schematic of the light-emitting microcavity device incorporating two identical STF chiral reflectors sandwiching a layer of ALq3 molecules. Here, θ denotes the emission cone. The oblique arrows drawn inside the layers simply underscore the back and force bouncing of photons.

 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.

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



Chiral STF

$$\underline{\epsilon}_{ref}^{o}(\omega) = \underline{\epsilon}_{ref}(\omega) \Big|_{\chi=0} = \epsilon_{a}(\omega) \mathbf{u}_{z} \mathbf{u}_{z} + \epsilon_{b}(\omega) \mathbf{u}_{x} \mathbf{u}_{x} + \epsilon_{c}(\omega) \mathbf{u}_{y} \mathbf{u}_{y}$$

$$\epsilon_a = 2.5 (1 + i\delta_{\epsilon}), \ \epsilon_b = 3.2 (1 + i\delta_{\epsilon}), \ \epsilon_c = 2.6 (1 + i\delta_{\epsilon})$$

 $\delta_\epsilon > 0$ absorption $\delta_\epsilon < 0 ~{\rm gain}$

 $\delta_\epsilon=0$ no absorption, no gain

Solve Maxwell postulates for reflection and transmission





High Density of States

implies

High Emission

Analogy: Lasing by dye-doped CLCs

$$\underline{\epsilon}_{PE}^{-1} = \begin{pmatrix} 1/\epsilon_1^{(0)} + \sum_{K=1}^3 r_{1K} E_K^{dc} & \sum_{K=1}^3 r_{6K} E_K^{dc} & \sum_{K=1}^3 r_{5K} E_K^{dc} \\ \sum_{K=1}^3 r_{6K} E_K^{dc} & 1/\epsilon_2^{(0)} + \sum_{K=1}^3 r_{2K} E_K^{dc} & \sum_{K=1}^3 r_{4K} E_K^{dc} \\ \sum_{K=1}^3 r_{5K} E_K^{dc} & \sum_{K=1}^3 r_{4K} E_K^{dc} & 1/\epsilon_3^{(0)} + \sum_{K=1}^3 r_{3K} E_K^{dc} \end{pmatrix}$$

$$\underline{\underline{\epsilon}}_{ref}^{o} = \underline{\underline{\epsilon}}_{PE}$$

DC voltage across the thickness



Available online at www.sciencedirect.com



Optics Communications

Optics Communications 259 (2006) 164-173

www.elsevier.com/locate/optcom

Electrically controlled optical bandgap in a structurally chiral material

J. Adrian Reyes ^{a,b}, Akhlesh Lakhtakia ^{b,c,*}

 ^a Instituto de Fisica, Universidad Nacional Autonoma de Mexico, Apartado 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:
(1) Reyes & Lakhtakia, *Opt. Commun.*, at press
(2) Lakhtakia & Reyes, *Phys. Rev. E*, submitted



 $\mathsf{R}_{_{\mathsf{RR}}}$ 1.0₁ $V_{dc} = 0$ $R_{RL} = R_{LR}$ 0.8 ----- R_{LL} Pseudo-0.6 Isotropic 0.4 point 0.2-0.0 1.7 1.8 λ 0.6 ⊢ 0.4 0.2 0.0 1.7 1.8 1.6 1.9 С λ

Without dc voltage



Without dc voltage

With dc voltage



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

Innovations in Materials Research, Vol. 1, No. 2 (1996) 165-176 © 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:

Harris, Westra, Brett, Electrochem. Sol.-St. Lett. 4, C39 (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)



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

Helical holes

Excellent for piezoelectrically controlled STFs



JOURNAL OF MODERN OPTICS, 2003, VOL. 50, NO. 2, 239-249

On piezoelectric control of the optical response of sculptured thin films

FEI WANG, AKHLESH LAKHTAKIA¹ and RUSSELL MESSIER

POLYMERIC STFs: REPLAMINEFORM TECHNIQUE (5-step)

INSTITUTE OF PHYSICS PUBLISHING

JOURNAL OF MICRONICHARICS AND MCROINGINGERING

J. Micromech. Microang. 15 (2005) 49-54

doi:10.1088/0960-1317/15/1/008

Large-area microfabrication of three-dimensional, helical polymer structures

A L Elias¹, K D Harris², C W M Bastiaansen², D J Broer^{2,3} and M J Brett¹

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





Polymer 46 (2005) 9544-9548

polymer

www.elsevier.com/locate/polymer

Polymer Communication Growth of sculptured polymer submicronwire assemblies by vapor deposition

Sean Pursel, Mark W. Horn, Melik C. Demirel*, Akhlesh Lakhtakia

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.



POLYMERIC STFs: HOLOGRAPHIC LITHOGRAPHY



Available online at www.sciencedirect.com





Photonics and Nanostructures - Fundamentals and Applications 3 (2005) 79-83

www.elsevier.com/locate/photonics

Photonic crystals with a chiral basis by holographic lithography E.R. Dedman^a, D.N. Sharp^{a,*}, A.J. Turberfield^a, C.F. Blanford^b, R.G. Denning^b

- 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



Horn, M.W., Pickett, M.D., Messier, R., Lakhtakia, A., NANOTECHNOLOGY, Vol. 15, pp. 303-310, 2004

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.









6. STFs WITH TRANSVERSE ARCHITECTURE

STFs WITH TRANSVERSE ARCHITECTURE

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







STFs WITH TRANSVERSE ARCHITECTURE



Chromium



Metal STFs on Topography



Molybdenum



Aluminum



STFs WITH TRANSVERSE ARCHITECTURE

Semiconductor STFs on Micro and Nanoscale Topography



SnO_x STFs grown on photoresist patterns
STFs WITH TRANSVERSE ARCHITECTURE

Sculptured Nanowires on Nanoscale Topography



Single SiOx nanowire array grown on 60 nm ebeam resist



BCC array of SiOx nanocolumns



HCP array of SiOx nanocolumns



1um x 1um mesh of SiOx nanolines

STFs WITH TRANSVERSE ARCHITECTURE

INSTITUTE OF PHYSICS PUBLISHING

Nanotechnology 15 (2004) 303-310

NAMOTECHNOLOGY

PII: S0957-4484(04)69259-2

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

Mark W Horn, Matthew D Pickett, Russell Messier and Akhlesh Lakhtakia¹

Selective growth of sculptured nanowires on microlithographic lattices

Mark W. Horn,^{a)} 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