On Percolation and Circular Bragg Phenomenon in Metallic, Helicoidally Periodic, Sculptured Thin Films

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Abstract: The concept of local homogenization is invoked in the visible and the sub-visible frequency regimes to estimate the permittivity dyadics of a thin-film helicoidal bianisotropic mediums (TFHBMs) — a canonical class of sculptured thin films with helicoidally periodic microstructure — made of a metal. Examination of the components of the permittivity dyadics, as well as of the co- and the cross-polarized reflectances of axially excited, metallic TFHBM halfspaces, reveals that three different absorbing-dielectric-to-metal transitions will occur as the metallic volumetric fraction is increased, but only two will be optically sensed using normally incident plane waves. The first transition is show to virtually eliminate the circular Bragg phenomenon characteristically displayed by axially excited, dielectric TFHBMs.

Keywords: Percolation; Circular Bragg phenomenon; Helicoidally periodic mediums; Sculptured thin films

1 Introduction

Microstructurally, a sculptured thin film (STF) is best visualized as an assembly of parallel columns that all curve in the same way in the thickness direction [1]. Thus, a STF is locally anisotropic and unidirectionally nonhomogeneous, and may be considered as a nonhomogeneous continuum in the visible and the sub-visible frequency regimes. The aim of this communication is to theoretically delineate the effect of percolation on the circular Bragg phenomenon (CBP) displayed by the so-called thin-film helicoidal bianisotropic mediums (TFHBMs) — a canonical class of STFs with helicoidally periodic microstructure — made of metals.

Metal-dielectric particulate composite materials are known to demonstrate the phenomenon of percolation [2] in the following manner: An insulator-to-conductor transition occurs abruptly as \( f \), the volumetric fraction of metallic particles, increases from zero. Provided the metallic particles are spherical, the transition occurs at \( f = f_{pt} \sim 0.33 \), where \( f_{pt} \) is the percolation threshold. Different values of \( f_{pt} \) are found for nonspherical particles, because it is a strong function of particulate geometry and orientational statistics. This was exemplified by Lakhtakia et al. [3] for uniaxial composite mediums formed by randomly dispersing parallel, aciculate (i.e., needle-like), dielectric particles in some isotropic dielectric host medium.

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One of the two Bruggeman models set up for the homogenization of uniaxial composite mediums [3] is also the basis of a simple model to predict the effective constitutive properties of dielectric TFHBMs [4], wherein the continuous columns are modeled as ensembles of needles. It stands to reason that percolation must be exhibited by metallic TFHBMs. Although a few metallic TFHBMs have been fabricated [5,6], their optical responses remain unexplored. The lack of experimental data may be due to the structural irregularities in early specimen of metallic TFHBMs [5], the causes of and the remedies for irregularities being topics of current research [6]- [8]. These laboratory fabrication efforts motivated the theoretical study reported here.

The plan of this paper is as follows: Section 2 contains the theoretical preliminaries. The permittivity dyadic of a metallic TFHBM is presented, and a simple homogenization model to estimate the relative permittivity scalars is briefly recounted. In Section 3, a boundary value problem is formulated for the reflection of a normally incident plane wave that excites a metallic TFHBM halfspace along its axis of helicoidal periodicity. The effect of percolation on the planewave response of the metallic TFHBM halfspace is addressed in Section 3. Vectors are underlined, dyadics are double-underlined; while an \( \exp(-i\omega t) \) time-dependence is implicit, with \( \omega \) as the angular frequency.

2 Theoretical Preliminaries

2.1 Permittivity dyadic

Although all previous investigations on STFs considered the reflection/transmission responses of films of infinite lateral extent and finite width, determination of the response of a metallic TFHBM halfspace is more appropriate in the present context. This is because excessive attenuation inside a TFHBM layer of finite width leads to virtually zero fields on the layer’s back face. The exact solution of a relevant two-point boundary value problem [9] is then severely compromised for even a moderately thick TFHBM layer.

Suppose the halfspace \( z \geq 0 \) is occupied by a metallic TFHBM, while the halfspace \( z \leq 0 \) is vacuous. The linear dielectric properties of the TFHBM are delineated by the nonhomogeneous permittivity dyadic

\[
\varepsilon(z) = \varepsilon_0 \cdot \mathbf{S}_z(z, h) \cdot \mathbf{S}_y(\chi) \cdot \varepsilon_{\text{ref}} \cdot \mathbf{S}_y^{-1}(\chi) \cdot \mathbf{S}_z^{-1}(z, h); \quad z \geq 0 ,
\]

where

\[
\varepsilon_{\text{ref}} = \varepsilon_a \mathbf{u}_x \mathbf{u}_x + \varepsilon_b \mathbf{u}_x \mathbf{u}_x + \varepsilon_c \mathbf{u}_y \mathbf{u}_y
\]

is called the local relative permittivity dyadic. The relative permittivity scalars \( \varepsilon_{a,b,c} \) are implicit functions of frequency, and the presented analysis applies to dielectric TFHBMs too. Here and hereafter, \( \varepsilon_0 = 8.854 \times 10^{-12} \text{ F/m} \) and \( \mu_0 = 4\pi \times 10^{-7} \text{ H/m} \) are the permittivity and the permeability of free space (i.e., vacuum), respectively; \( k_0 = 2\pi / \lambda_0 = \omega \sqrt{\mu_0 \varepsilon_0} \) and \( \lambda_0 \) are the wavenumber and wavelength in free space, respectively; \( \eta_0 = \sqrt{\mu_0 / \varepsilon_0} \) is the intrinsic impedance of free space; while \( \mathbf{u}_x, \mathbf{u}_y \) and \( \mathbf{u}_z \) denote the unit vectors in a cartesian coordinate system.
The helicoidal periodicity of the chosen TFHBM is captured by the rotation dyadic

\[ S_z(z,h) = u_z u_z + (u_y u_y + u_y u_y) \cos \frac{\pi z}{\Omega} + h(u_y u_x - u_x u_y) \sin \frac{\pi z}{\Omega}. \]  

(3)

The direction of the nonhomogeneity is parallel to the \( z \) axis, with \( 2\Omega \) as the structural period. The integer \( h = 1 \) for a structurally right-handed TFHBM; and \( h = -1 \) for structural left-handedness.

The tilt dyadic

\[ S_y(\chi) = u_y u_y + (u_x u_x + u_y u_z) \cos \chi + (u_z u_x - u_x u_z) \sin \chi. \]  

(4)

represents the locally aciculate microstructure of a TFHBM, with \( \chi > 0^\circ \) being the tilt angle [4,7].

### 2.2 Estimation of \( \epsilon_{a,b,c} \)

Let the chosen TFHBM be fabricated of an isotropic material whose relative permittivity in the bulk state is denoted by \( \epsilon_s \) at the frequency of interest. The columns are supposed to comprise identical long needles, whose length is \( u (\gg 1) \) times greater than their cross-sectional radius. Additionally, without loss of generality in the present context, the cross-section of the needles is assumed to be circular, which implies that \( \epsilon_c = \epsilon_a \) in (2). The needles are electrically small, the film is porous, and the void regions are vacuous. The orientation of the needles changes with \( z \), but not with \( x \) and \( y \). The TFHBM can thus be viewed locally as a two-phase composite material with the so-called 3-1 connectivity [10], as the vacuous phase is connected in 3-D and the metallic phase in 1-D. Of course, the 1-D connectivity of the metallic phase twists with \( z \), in accordance with the helix described by the unit vector

\[ u_t(z) = S_z(z,h) \cdot S_y(\chi) \cdot u_x. \]  

(5)

The concept of local homogenization [4] is invoked now to estimate the effective dielectric response properties of the chosen TFHBM. Application of the Bruggeman formalism leads to the dyadic equation [3,4]

\[ \begin{align*}
0 & = (1-f) \left[ \epsilon_{ref}^o - I \right] \cdot \left\{ I - W \cdot \left[ I - \epsilon_{ref}^o \epsilon_s \right]^{-1} \right\}^{-1} \\
& + f \left[ \epsilon_{ref}^o - \epsilon_s I \right] \cdot \left\{ I - W \cdot \left[ I - \epsilon_s \epsilon_{ref}^o \epsilon_s \right]^{-1} \right\}^{-1},
\end{align*} \]  

(6)

where the shape factor \( u \) of the needles occurs in the dyadic

\[ W = \frac{1}{2} \left[ 1 + 4u^{-2} \frac{\epsilon_b}{\epsilon_a} \right]^{-1/2} I + \left\{ 1 - \frac{3}{2} \left[ 1 + 4u^{-2} \frac{\epsilon_b}{\epsilon_a} \right]^{-1/2} \right\} u_x u_x; \]  

(7)

the metallic volumetric fraction is denoted by \( f \), \( 0 \leq f \leq 1 \); while \( I \) and \( 0 \) are the unit and the null dyadics, respectively. Equation (6) has to be solved numerically to estimate the constituents \( \epsilon_a \) and \( \epsilon_b \) of \( \epsilon_{ref}^o \).
3 Boundary Value Problem

Suppose an arbitrarily polarized plane wave is normally incident on the chosen TFHBM halfspace from the lower halfspace \( z \leq 0 \). As a result of the axial excitation of the upper halfspace, a plane wave is reflected into the lower halfspace. The electric field phasor associated with the two plane waves in the lower halfspace is stated as

\[
\mathbf{E}(z) = \left( a_L \mathbf{u}_+ + a_R \mathbf{u}_- \right) \exp(ik_0z) + \left( r_L \mathbf{u}_- + r_R \mathbf{u}_+ \right) \exp(-ik_0z) ; \quad z \leq 0 ,
\]

and the corresponding magnetic field phasor is then easily determined from the Faraday equation. Here, the complex unit vectors \( \mathbf{u}_\pm = (\mathbf{u}_x \pm i\mathbf{u}_y)/\sqrt{2} \); \( a_L \) and \( a_R \) are the known amplitudes of the left– and the right–circularly polarized (LCP & RCP) components of the incident plane wave; and \( r_L \) and \( r_R \) are the unknown amplitudes of the reflected planewave components. Our intention is to determine the reflection coefficients entering the \( 2 \times 2 \) matrix in the following relation:

\[
\begin{bmatrix}
    r_L \\
    r_R 
\end{bmatrix} =
\begin{bmatrix}
    r_{LL} & r_{LR} \\
    r_{RL} & r_{RR} 
\end{bmatrix}
\begin{bmatrix}
    a_L \\
    a_R 
\end{bmatrix} .
\]

These coefficients are doubly subscripted: those with both subscripts identical refer to co-polarized, while those with two different subscripts denote cross-polarized, reflection.

The specification of fields induced in the TFHBM halfspace requires some care. Four modes can propagate in the \( \pm z \) direction; therefore

\[
\mathbf{E}(z) = \sum_{n=1}^{4} a_n e^{ig_nz} \left( \mathbf{u}_x [e_{n1} \cos(\pi z/\Omega) - e_{n2} \sin(\pi z/\Omega)] \\
+ \mathbf{u}_y [e_{n1} \sin(\pi z/\Omega) + e_{n2} \cos(\pi z/\Omega)] + \mathbf{u}_z e_{n3} \right) ; \quad z \geq 0 ,
\]

and

\[
\mathbf{H}(z) = \sum_{n=1}^{4} a_n e^{ig_nz} \left( \mathbf{u}_x [h_{n1} \cos(\pi z/\Omega) - h_{n2} \sin(\pi z/\Omega)] \\
+ \mathbf{u}_y [h_{n1} \sin(\pi z/\Omega) + h_{n2} \cos(\pi z/\Omega)] \right) ; \quad z \geq 0 .
\]

The (un–normalized) cartesian components of the modal field phasors, given by [9,11]

\[
\begin{align*}
    e_{n1} &= \omega \mu_0 \left[ g_n^2 - k_0^2 \varepsilon_c + \left( \frac{\pi}{\Omega} \right)^2 \right] \\
    e_{n2} &= 2i \omega \mu_0 \frac{\pi}{\Omega} g_n \\
    e_{n3} &= e_{n1} \frac{\varepsilon_a - \varepsilon_b}{\varepsilon_a \varepsilon_b} \tilde{\varepsilon}_d \cos \chi \sin \chi \\
    h_{n1} &= -i \frac{\pi}{\Omega} \left[ g_n^2 + k_0^2 \varepsilon_c - \left( \frac{\pi}{\Omega} \right)^2 \right] \\
    h_{n2} &= g_n \left[ g_n^2 - k_0^2 \varepsilon_c - \left( \frac{\pi}{\Omega} \right)^2 \right] 
\end{align*}
\]

contain the four modal wavenumbers

\[
g_1 = -g_3 = +2^{-1/2} \left\{ k_0^2 (\varepsilon_c + \tilde{\varepsilon}_d) + 2 \left( \frac{\pi}{\Omega} \right)^2 \\
+ k_0 \left[ k_0^2 (\varepsilon_c - \tilde{\varepsilon}_d)^2 + 8 \left( \frac{\pi}{\Omega} \right)^2 (\varepsilon_c + \tilde{\varepsilon}_d) \right] \right\}^{1/2} ,
\]

(13)
\[ g_2 = -g_4 = +2^{-1/2} \left\{ k_0^2 (e_c + \tilde{\epsilon}_d) + 2 \left( \frac{\pi}{\Pi} \right)^2 \right. \]
\[ - k_0 \left[ k_0^2 (e_c - \tilde{\epsilon}_d)^2 + 8 \left( \frac{\pi}{\Pi} \right)^2 (e_c + \tilde{\epsilon}_d)^{1/2} \right]^{1/2}; \]

while
\[ \tilde{\epsilon}_d = \frac{\epsilon_a \epsilon_b}{\epsilon_a \cos^2 \chi + \epsilon_b \sin^2 \chi} \]
is defined for convenience. We assume here that \( \Omega \) is finite and exclude the possibility of excitation of axially propagating Voigt waves [12].

The summation symbols in (10) and (11) are primed to indicate that two of the four modal coefficients \( a_n \), \( 1 \leq n \leq 4 \), must be identically null-valued in the present context. The determination of which two requires analysis (and computation) of the \( z \)-directed components of the modal time-averaged Poynting vectors \( P_n(z) \), \( 1 \leq n \leq 4 \); thus,

\[ P_{nz}(z) = \frac{1}{2} |a_n|^2 \exp \{ -2 \text{Im}[g_n] z \} \text{Re} \left[ e_{n1} h_{n2}^* - e_{n2} h_{n1}^* \right]; \quad 1 \leq n \leq 4, \]

where the asterisk denotes the complex conjugate. Because \( P_{1z} > 0 \) and \( P_{3z} < 0 \), in general [11], we must have \( a_1 \neq 0 \) and \( a_3 \equiv 0 \). The quantities \( P_{2z} \) and \( P_{4z} \) are always opposite in sign; hence, either \( a_2 \equiv 0 \) when \( P_{2z}/|a_2|^2 < 0 \), or \( a_3 \equiv 0 \) when \( P_{4z}/|a_4|^2 < 0 \). This process also ensured, for all calculations presented here, that \( \text{Im}[g_1] \geq 0 \) as well as that either \( \text{Im}[g_2] \geq 0 \) or \( \text{Im}[g_4] \geq 0 \), as appropriate.

The boundary value problem can now be formulated by ensuring the continuity of the tangential components of the electric and the magnetic field phasors across the plane \( z = 0 \). The following four algebraic equations emerge:

\[ (a_L + a_R) + (r_L + r_R) = \sqrt{2} \sum _{n=1}^{4} \prime a_n e_{n1}, \]
\[ (a_L - a_R) - (r_L - r_R) = -\sqrt{2} i \sum _{n=1}^{4} \prime a_n e_{n2}, \]
\[ (a_L - a_R) + (r_L - r_R) = \sqrt{2} i \eta_0 \sum _{n=1}^{4} \prime a_n h_{n1}, \]
\[ (a_L + a_R) - (r_L + r_R) = \sqrt{2} i \eta_0 \sum _{n=1}^{4} \prime a_n h_{n2}. \]

Their solution yields the four coefficients \( r_{RR} \), etc.

4 Numerical Results and Discussion

All four reflection coefficients were computed using Mathematica 3.0 on a Power Macintosh 7300/180 computer. The values \( h = 1 \), \( \lambda_0 = 600 \text{ nm} \), \( \chi = 30^\circ \) and \( u = 10 \) were fixed for all calculations;
while the volumetric fraction \( f \) and the structural half-period \( \Omega \) were varied. For a specified \( \epsilon_s \), first \( \epsilon_a = \epsilon_c \) and \( \epsilon_b \) were computed by solving (6) iteratively; and then the reflection coefficients \( r_L \) and \( r_R \) were obtained from the simultaneous solution of (17)-(20). The relative permittivity scalars \( \epsilon_a \) and \( \epsilon_b \) were plotted as functions of \( f \), and the reflectances

\[
R_{pq} = |r_{pq}|^2; \quad p = L, R; \quad q = L, R, \tag{21}
\]

as functions of both \( f \) and \( \Omega \).

In order to understand the response of a metallic TFHBM halfspace, it is best to begin with the results for a virtually lossless dielectric TFHBM halfspace. Shown in Figure 1 are the computed values of \( R_{RR} \), \( R_{LL} \) and \( R_{LR} = R_{RL} \) — along with \( \epsilon_a \) and \( \epsilon_b \) — when \( \epsilon_s = 5 + i0.001 \). The TFHBM chosen is structurally right–handed; and the CBP is manifested (i) as the very high-magnitude ridge in the plot of \( R_{RR} \) as well as (ii) the absence of that feature in the plot of \( R_{LL} \). This manifestation had been observed earlier for axially excited, lossless dielectric TFHBM layers [9,13], but not for halfspaces. The CBP vanishes when either \( f = 0 \) or \( f = 1 \), because the TFHBM is then isotropic (i.e., \( \epsilon_a = \epsilon_b = \epsilon_c \)) and, therefore, homogeneous.

Next let us delineate the effect of absorption. The calculations for Figure 1 were repeated, but with \( \epsilon_s = 5 + i0.5 \). As the volumetric fraction \( f \) increases, absorption becomes increasingly significant, as shown by the plots of \( \epsilon_a \) and \( \epsilon_b \) in Figure 2. The CBP still leaves a broadly recognizable signature — as also for absorbing TFHBM layers [14] — but the initially high-magnitude ridge in the plot of \( R_{RR} \) dissipates fairly rapidly as \( f \) increases from zero. Indeed, the CBP virtually disappears for \( f \gtrsim 0.8 \).

Finally, in Figure 3 are shown the calculated results for a metallic TFHBM halfspace. The chosen value \( \epsilon_s = -50 + i18 \) is very close to the relative permittivity of bulk aluminum at \( \lambda_0 = 600 \text{ nm} \) [15]. The CBP has a very clear signature in the plot of \( R_{RR} \) in Figure 3, but only for very low values of \( f \), when the TFHBM is effectively an absorbing dielectric material. The high-magnitude ridge broadens relative to the one in Figure 2, and vanishes for \( f \gtrsim 0.4 \). The reason is the occurrence of percolation in the two-phase composite material that the chosen TFHBM is [10].

Two absorbing-dielectric-to-metal transitions are evident in the plots of \( \epsilon_a \) and \( \epsilon_b \) in Figure 3. The first transition occurs at the low value \( f \sim 0.05 \), \( \text{Re}[\epsilon_b] \) becoming negative thereafter and \( \text{Im}[\epsilon_b] \) increasing in magnitude as \( f \) increases thereafter. The second transition occurs at \( f \sim 0.5 \), with \( \text{Re}[\epsilon_a] < 0 \) thereafter and \( \text{Im}[\epsilon_a] \) increasing in magnitude as \( f \) increases thereafter.

The reason for the two transitions not coinciding becomes clear on rewriting (1) and (2) together as

\[
\underline{\epsilon}(\nu) = \epsilon_0 \left\{ \epsilon_a \left[ L - \underline{u}_l(z)\underline{u}_l(z) \right] + \epsilon_b \underline{u}_l(z)\underline{u}_l(z) \right\}, \tag{22}
\]

wherein the equality \( \epsilon_c = \epsilon_a \) has been assumed. Percolation along the direction parallel to \( \underline{u}_l(z) \) occurs at lower threshold values of \( f \) because of the connectedness of the metallic needles in that direction; whereas the lower connectivity of the metallic phase in all other directions delays the onset of percolation in those directions. As \( f \) increases, percolation occurs last in any direction perpendicular to \( \underline{u}_l(z) \). In Figure 3, the inter-transition \( f \)-regime is most clearly evident as \( 0.4 \lesssim f \lesssim 0.6 \) in the plot of \( R_{LR} \), which becomes virtually constant at a high magnitude for \( f \gtrsim 0.6 \).
Although only two different absorbing-dielectric-to-metal transitions are evident in the plots of $\epsilon_a$ and $\epsilon_b$ in Figure 3 because of the assumption that $\epsilon_c = \epsilon_a$, three different transitions will occur for a metallic TFHBM for which $\epsilon_c \neq \epsilon_a$. As $f$ will increase from 0, the first transition will be exhibited by $\epsilon_b$. The second and the third transitions will be located at higher values of $f$, in consequence of the columnar microstructure being locally aciculate. The latter two transitions will coincide for columns with circular cross-sections, but not for noncircular columns. However, the reflection of normally incident plane waves by metallic TFHBM halfspaces and layers shall show evidence of only two transitions, because only (i) $\epsilon_c$ and (ii) the combination $\tilde{\epsilon}_d$ of $\epsilon_a$ and $\epsilon_b$ enter the expressions (10)-(14). We also conclude from the presented results that metallic TFHBMs with only low metallic volumetric fractions appear desirable, if the aim is to exploit the CBP for a certain application.
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Lakhtakia, Figure 1

Figure 1: Computed values of $R_{RR}$, $R_{LL}$ and $R_{LR} = R_{RL}$ as functions of $f$ and $\Omega$; as well as computed values of $\epsilon_a = \epsilon_c$ and $\epsilon_b$ as functions of $f$. The TFHBM halfspace is made of a virtually lossless dielectric material with $\epsilon_s = 5 + i0.001$, so that $\text{Im}[\epsilon_a]$ and $\text{Im}[\epsilon_b]$ are too small to be shown. Other parameters are as follows: $h = 1$, $\lambda_0 = 600$ nm, $\chi = 30^\circ$ and $u = 10$. 
Figure 2: Same as Figure 1, but for a TFHBM halfspace made of an absorbing dielectric material with $\epsilon_s = 5 + i0.5$. 

Lakhtakia, Figure 2
Lakhtakia, Figure 3

Figure 3: Same as Figure 1, but for a TFHBM halfspace made of a metal with $\epsilon_s = -50 + i18$. 