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Jason C. Vap

Stephen E. Nauyoks
Air Force Institute of Technology

Michael R. Benson

Michael A. Marciniak
Air Force Institute of Technology

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Use of a novel infrared wavelength-tunable laser Mueller-matrix polarimetric scatterometer to measure nanostructured optical materials

Jason C. Vap,¹,² Stephen E. Nauyoks,³,⁴ Michael R. Benson,³,⁵ and Michael A. Marciniak³

¹Department of Electrical and Computer Engineering, Air Force Institute of Technology, Wright-Patterson AFB OH 45433, USA
²846 Test Squadron, Holloman AFB NM 88330, USA
³Department of Engineering Physics, Air Force Institute of Technology, Wright-Patterson AFB OH 45433, USA
⁴Oak Ridge Institute of Science and Education, Belcamp MD 21017, USA
⁵Materials and Manufacturing Directorate Air Force Research Laboratory, Wright-Patterson AFB OH 45433, USA

Nanostructured optical materials, for example metamaterials, have unique spectral, directional and polarimetric properties. Samples designed and fabricated for infrared (IR) wavelengths have been characterized using broad-band instruments to measure specular polarimetric transmittance or reflectance as in ellipsometry, or integrated hemisphere transmittance or reflectance. We have developed a wavelength-tunable IR Mueller-matrix (Mm) polarimetric scatterometer which uses tunable external-cavity quantum-cascade lasers (EC-QCL’s) to tune onto and off of the narrow-band spectral resonances of nanostructured optical materials, and perform full polarimetric and directional evaluation to more fully characterize their behavior. Using a series of EC-QCL’s the instrument is tunable over 4.37-6.54 μm wavelengths in the mid-wave IR and 7.41-9.71 μm in the long-wave IR, and makes measurements both at specular angles, acting as a Mm polarimeter, and off-specular, acting as a Mm scatterometer. Example measurements of an IR thermal metamaterial are shown.

I. INTRODUCTION

Modern optical materials may be designed to have unique spectral, directional and polarimetric optical properties. We have developed a unique, spectrally tunable, fully Stokes-polarimetric scatterometry system in the infrared (IR) specifically for the characterization of nanostructured optical materials, e.g., photonic crystals, plasmonic structures and metamaterials.¹⁴ Nanostructured IR materials offer unique measurement challenges. Theoretical predictions of optical metamaterials have shown that the effective material parameters of interest, electric permittivity (ε) and magnetic permeability (μ), will often depend on the incident angle and polarization of the incident radiation.⁵,⁶ Also, the attribution of optical phenomena observed or predicted for these materials, e.g., to Bragg resonance as opposed to constitutive, homogeneous (effective) material parameters, based on inclusion size and periodicity, may not be clear cut.⁷,⁸ Finally, these optical phenomena, often based on Bragg or plasmonic resonances, are typically narrow band in nature.⁹,¹⁰ Past characterization of fabricated visible/IR optical nanostructured materials has included Fourier-transform-spectrometric specular transmittance¹¹-¹⁴ and reflectance,¹⁵-¹⁷ and emittance,¹⁸-²⁶ IR spectrophotometric specular transmittance and reflectance,²⁷,²⁸ IR prism-spectrometer emittance,²⁹,³⁰ Variable-Angle Spectrometric Ellipsometry (VASE),⁹ and hemispheric absorptance measured with an integrating sphere.³¹,³² However, the capability to perform optical characterization of
nanostructured material samples which is complete from a spectral, polarization and directional point of view has not been available to IR nanostructured materials researchers.

Narrow-band performance features of nanostructured optical materials are often observed using a broadband instrument. A spectrometer typically provides reflectance and transmittance data at normal incidence or fixed, specular reflectance or transmittance angles. When increasing the measurement space to include polarization-sensitive measurements, a VASE also provides data at specular angles. Further, the IR-VASE typically provides a limited polarimetric evaluation, i.e. co- and cross-polarization (s- (TE) polarization incident/s-polarization reflected, pp (TM/TM), sp or ps) measurements and only partial Mueller-matrix (Mm) extractions. For fully polarimetric characterization at IR wavelengths both at and away from specular (and both in and out of the plane of incidence), an IR Mm polarimetric scatterometer is required. However, typical scatterometers are often limited in their ability to characterize nanostructured optical materials by their fixed-wavelength laser sources.

We have incorporated a series of IR-tunable external-cavity quantum-cascade lasers (EC-QCL’s) to address this. Six tunable Daylight Solutions® EC-QCL’s were selected to span a nearly continuous range of mid-wave IR (MWIR, 4.37-6.54-μm) and long-wave IR (LWIR, 7.41-9.71-μm) wavelengths, and added to two fixed-wavelength 3.39-μm HeNe and 10.6-μm CO2 lasers in a Schmitt Measurement Systems (SMS) Complete Angle Scatter Instrument (CASI®). Extensive hardware and software upgrades were also implemented to introduce an achromatic Dual Rotating Retarder (DRR) polarimeter with associated automated rotation stages to this instrument. The end product is a spectrally tunable IR Mm polarimetric scatterometer, which can be tuned into and out of narrow-band performance regions of nanostructured optical materials, allowing for the investigation of their full range of spectral, directional and polarimetric behaviors.

II. INSTRUMENT

A. General scatterometer layout

The CASI® already had an existing and suitable beam train for the introduction of additional laser sources, but modifications were made so the EC-QCL’s could be introduced and easily aligned. Figure 1 shows turning mirrors (TM’s) and a beam combiner (BC) to couple EC-QCL’s into the beam path. As importantly, they also co-align a visible laser into the same beam path to facilitate initial system alignment. Alignment is also facilitated by power adjustments to the EC-QCL, where thermal paper is used to trace the IR beam through the system from source to detector. Careful alignment is required to locate the IR beam on the front surface of the sample at the center of rotation of the goniometer (right side of Figure 1, where the incident, \( \theta_i \), and scatter, \( \theta_s \), angles are measured from sample normal). \( \theta_i \) and \( \theta_s \) are measured accurately only after this careful alignment.
Figure 1. Overhead-view schematic of the tunable Mm polarimetric scatterometer. IR-source and visible-alignment lasers are aligned along the optical path using turning mirrors (TM) and a beam combiner (BC). The beam is chopped by a chopper (Ch) to allow lock-in detection with a lock-in amplifier (Li). The beam is focused through a pinhole (PH) by a focusing lens (FL), and the diverging beam focused through the sample location onto the detector by an off-axis parabolic (OAP) mirror to provide nearly collimated incidence on the sample. The pinhole provides background suppression for the detector looking back at the source. The sample mount is on the right at the center of rotation of a goniometer; incident ($\theta_i$) and scatter ($\theta_s$) angles are measured from sample normal ($\theta_n$). The detector is at the end of the goniometer arm, which moves independently of sample position. The sample may also be tipped about its $\theta_n$-axis for out-of-plane measurements. A Stokes polarization state is generated with a polarizer (GP)-retarder (GR) pair and analyzed with another retarder (AR)-polarizer (AP) pair.

B. Spectrally tunable IR sources

Six tunable EC-QCL’s span much of the MWIR and LWIR wavelengths. Table I shows the tunable range of each EC-QCL, their peak wavelengths and maximum powers. Unlike discharge lasers, the power of an EC-QCL is directly adjustable, which is effective for alignment, improvement of signal detection, and achieving the largest dynamic range possible for the instrument. The output powers of the 3.39-μm HeNe and 10.6-μm CO$_2$ lasers are 3.7 mW and 14.6 W, respectively.

<table>
<thead>
<tr>
<th>EC-QCL wavelength range (μm)</th>
<th>$\lambda_{\text{peak}}$ (μm)</th>
<th>Maximum Power (mW)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.35-4.55</td>
<td>4.5</td>
<td>220</td>
</tr>
<tr>
<td>4.74-5.12</td>
<td>4.9</td>
<td>160</td>
</tr>
<tr>
<td>5.16-5.67</td>
<td>5.3</td>
<td>330</td>
</tr>
<tr>
<td>5.76-6.54</td>
<td>6.1</td>
<td>280</td>
</tr>
<tr>
<td>7.40-8.23</td>
<td>7.8</td>
<td>340</td>
</tr>
<tr>
<td>8.06-9.71</td>
<td>8.8</td>
<td></td>
</tr>
</tbody>
</table>

The wavelength-tuning ranges of the sources were tested using a Bristol Instruments® 721B spectrum analyzer and found to have excellent wavelength stability with bias. The sample set of results shown in Table II demonstrates that the tuned-to-measured wavelength deviation is strongly correlated to the tuned wavelength rather than being dependent on the applied bias current. In general, the EC-QCL wavelength is able to be set to six significant-digit accuracy with relative uncertainty of less than 0.2%, and laser line-widths are less than 30 MHz continuous wave and 30 GHz pulsed.

<table>
<thead>
<tr>
<th>EC-QCL tuned wavelength</th>
<th>Measured wavelength</th>
<th>Deviation</th>
</tr>
</thead>
</table>

Table II. EC-QCL wavelength stability performance under different bias conditions.
Automated wavelength tuning of the EC-QCLs is not currently a capability of our instrument. As noted, the scatterometer requires stringent alignment, and the EC-QCLs grating-tune the wavelength, which creates enough beam walk that, when translated over the length of the optical path, requires manual readjustments to the alignment.

C. Achromatic polarimetric components

The most complex element of this instrument is the achromatic DRR polarimeter. A DRR polarimeter can determine the full \( \text{Mm} \) of a sample under test using a polarization state generator (GP and GR in Figure 1) before the sample and polarization state analyzer (AR and AP in Figure 1) after, provided the retarders (GR and AR) are not half-wave plates (\( \lambda/2 \) retarders). The two retarders rotate at different rates, producing a data set of generated/analyzed polarizations from which the \( \text{Mm} \) may be extracted. Up-front analysis was conducted to determine the optimal retardance configuration and the best AR:GR rotation combination,\(^3,4,8\) both of which leverage an existing calibration method.\(^33\) Low-error \( \text{Mm} \) extractions are only possible with a robust calibration methodology. We concluded that a (\( \theta_A: \theta_G = 5\omega:1\omega \)) Fourier rotation scheme,\(^34\) where \( \theta_A \) is the rotation increment of AR and \( \theta_G \) of GR, such that the AR angular rotation increments (\( \omega \)) are five times larger than those of GR, was optimal by applying condition number and error analysis.\(^4\) The traditional Fourier rotation scheme uses quarter-wave plates (\( \lambda/4 \) retarders), but we found that Chenault’s calibration methodology\(^33\) could also be used for our optimal \( \lambda/3 \) retardance configuration\(^35,36\) and was better.\(^4\)

Achromatic, nominally \( \lambda/3 \) CdS/CdSe wave-plates were obtained from Gooch and Housego LLC for both the MWIR and LWIR wavebands. Actual retardance values ranged 113-127° and 109-116° for the 4.3-6.5-\( \mu \)m and 7.4-10.6-\( \mu \)m bands, respectively. Four high-precision Aerotech, Inc. AGR-50 rotation stages were added and electronically integrated into the CASI® instrument to achieve the DRR configuration. The AGR-50 has an internal 50:1 angular reduction ratio giving a calculated, repeatable step size of 0.016° when driven by a 0.8° increment stepper motor.\(^1,2\) This is well within the 0.3° DRR accuracy recommended by Goldstein.\(^37\) Electronic drivers for the Aerotech stages of the DRR were installed and interfaced with the existing instrument software. Software modifications were made to produce a seamless transition from the typical scalar directional scans of the commercial CASI® to our polarimetric scans.\(^1,2\)

Using Fourier rotation schemes of (\( \theta_A: \theta_G = 25\omega:5\omega \)) and (37.5°:7.5°), free-space \( \text{Mm} \) extractions at less than 1% error are consistently achieved. An example free-space \( \text{Mm} \) extraction is

\[
M_{\text{free-space}} = \begin{bmatrix}
1.0000 & 0.0010 & 0.0016 & -0.0019 \\
-0.0003 & 0.9978 & -0.0053 & 0.0012 \\
-0.0002 & 0.0065 & 0.9951 & 0.0010 \\
-0.0027 & -0.0016 & -0.0051 & 0.9969
\end{bmatrix}, \quad (1)
\]
III. EXPERIMENTAL RESULTS AND ANALYSES

To illustrate the capabilities of our instrument, Mm results from a spectrally, directionally and polarimetrically selective IR metamaterial absorber (MMA) sample, similar to that reported by Liu,16 are shown in Sections III A and B with the instrument behaving first as a Mm polarimeter, then as a Mm scatterometer. The MMA sample is an array of gold crosses whose crossbars alternate between 2.0 and 3.2-μm in length and are 400-nm wide, for an overall unit cell dimension of 7-μm, on a dielectric background. For comparison a J.A. Woollam Co. IR-VASE was used to take reflectance measurements. Figure 2 shows the results of the IR-VASE measurements and clearly shows a resonant feature at 5.0-μm and no resonant feature at 4.4-μm. Using one EC-QCL tuned to 5.0-μm wavelength and another tuned to 4.4-μm, the impetus for building this instrument is clearly demonstrated; i.e. to investigate the polarimetric content of resonant and off-resonant narrow-band features of IR nanostructured optical materials both at and away from specular angles.

**Figure 2.** P-pol (left) and s-pol (right) reflectance measurements of the MMA taken with the IR-VASE with increasing incident angle.

A. Example spectral Mm polarimetric characterization

Our instrument was set to collect specular reflectance from the IR metamaterial absorber over a solid angle of 50 μSr (i.e. 4-mm aperture diameter at 50-cm distance) at incident angles ranging 15-65°, with 1° increments collected over 20-30° near the resonant condition of θi = 25°, and with 5° increments outside this region. This measurement was repeated twice to demonstrate the importance of spectral tunability. First the wavelength was tuned to 5.0-μm which corresponds to a spectral resonant feature which was dependent on the incident angle of the source. The instrument was then tuned to 4.4-μm where there was no observed resonant feature.

Figure 3 shows the Mm results in the traditional 4x4 Mm format but where each element is now a plot of the behavior of that element as a function of incident (and in this case equivalently reflectance) angle. The plot in the m00 position shows the first measurement collected at each incident angle, which is the co-polarization (i.e. p-polarization incident, p-polarization reflected) Bidirectional Reflectance Distribution Function (BRDF) value. In this case, the presence of the resonant feature in this IR metamaterial at θi = 25° is first observed in the m00 element at a wavelength of 5.0-μm (solid blue lines in Figure 3). It is more clearly observed in several of the other Mm elements (m01, m10, m23, m34, m32 and m33) at
the 5.0-μm wavelength and is clearly incident-angle dependent. Note that this feature is only accessible with the wavelength tunability and is not observable with the 4.4-μm measurement (dashed red lines).

Away from this resonant feature, the underlying Mm behavior indicates this metamaterial acts like a metal mirror at these wavelengths. The resonance incident angle of 25° is believed to be the phase-matching condition for a Surface Plasmon Polariton (SPP) mode in this nanostructure. At resonance, this metamaterial displays both polarizer and retarder characteristics. The polarizer characteristics are observed in the m_{00}, m_{01} and m_{10} elements, and show that the reflectance of incident p-polarized light at the wavelength of 5.0-μm is reduced significantly at this incident angle. The retarder characteristics are observed in the m_{22}, m_{32}, m_{23} and m_{33} elements, and show the phase of reflected s-polarized light at the 5.0-μm wavelength and 25° incidence angle lags that of the reflected p-polarized light.

Figure 3. Specular-reflectance Mm plot at 5.0-μm (solid blue line) and 4.4-μm (dashed red line) wavelengths for an IR MMA sample similar to that described in Liu16 when the instrument operates as a Mm polarimeter, i.e. in specular mode such that the angle in each Mm element represents both incident and reflected angles. Note the nanostructure’s spectrally, directionally and polarimetrically unique resonance at 5.0-μm is identified at 25° in several of the elements.
B. Example directional characterization

The same IR metamaterial was also examined with our instrument set as a Mm scatterometer. Figure 4 is again in Mm format, but now shows data collected at a wavelength of 5.0-μm and incident angles of (a) 25°, the observed resonant condition, and (b) 60°, an off-resonant angle. As a scatterometer, the instrument collects Mm BRDF, so the angles shown for each Mueller element represent the in-plane reflectance angle for that particular incident angle. In Figure 4 (a) at the specular angle of 25° (represented by 0° in each Mm element), the same information shown in Figure 3 at 25° is shown again here. Away from specular, diffraction orders are observed, indicating that this sample was fabricated in “tiles” approximately 140 μm in size (the mask size of this particular electron-beam lithography (EBL) process) and “tiled” together in a square lattice to form the larger sample. The polarization content of the light scattered by this sample is superimposed on this diffraction pattern. In Figure 4 (b), the information in Figure 3 at 60° (where there is no resonant feature) is again shown at 0° (again, specular). Diffraction orders similar to those of Figure 1 (a) and the superposition of polarimetric information on these orders are again observed.
Figure 4. Mm BRDF of an IR MMA sample similar to that described in Liu\textsuperscript{16} at 5.0-\textmu m wavelength and (a) 25\degree and (b) 60\degree incident angles. In (a), the resonant feature shown in Figure 2 is observed here at 0\degree (i.e., the specular angle). The Mm in (b) represents an off-resonance condition, and no unique features at specular (i.e., 0\degree) are observed. In both (a) and (b), the periodic structure observed is due to diffraction from a 140-\textmu m EBL periodicity which was superimposed on the 7-\textmu m IR unit cell periodicity during fabrication.
IV. CONCLUSION

Analyses of the spectral, directional and polarimetric behaviors of narrow-band resonant features observed in nanostructured optical materials have previously been constrained by the limited data available from broadband instruments such as spectrometers used primarily at specular angles in reflectance or transmittance, and under limited polarimetric conditions. We have developed a tunable IR Mm polarimetric scatterometer to more fully explore the narrow-band features of nanostructured optical materials, both at and away from specular, and both in and out of the plane of incidence. Significant up-front analysis allowed this instrument to be designed and built in an optimal DRR polarimeter configuration, and the free-space results demonstrate excellent calibration performance. The example measurements of an IR metamaterial selective absorber demonstrate this instrument’s unique capability to provide novel spectral, directional and polarimetric information on such samples.

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