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Metamaterial metal-dielectric photonic bandgap filters with high in-band transmittance and strong out-of-band rejection (grant #FA9550-09-1-0418)

Principal Investigator: **Bernard Kippelen,** *Professor*

This program aimed to make advances in the design and fabrication of metamaterial metal-dielectric photonic bandgap (MDPBGs) linear optical filters. The goal was to deliver MDPBGs optical filters that provided stronger and wider out-of-band electromagnetic shielding than conventional multilayer dielectric "hot mirror" filters and, larger and wider transmittance passbands than current MDPBG linear optical filters. The objectives of this program were to:

1) investigate the influence of transition metal seeding on the optical properties, surface morphology and electrochemical stability of thin Ag layers deposited on high and moderate refractive index metal oxides.
2) identify a suitable set of dielectrics that enables the fabrication of single metal layer and multilayer metal-dielectric structures containing electrically continuous Ag layers with thicknesses ranging from 10 to around 20 nm.
3) investigate novel MDPBG optical filter designs that allow control over the spectral characteristics of its passband.
4) design and fabricate MDPBG linear optical filters that achieve spectrally broadband passbands ($\Delta\lambda_{FHWM}$>300 nm) with high transmittance (T > 60 %) across the visible spectral region (400 – 700 nm), strong out-of-band rejection up to radio frequencies, and good environmental stability.

Final Technical Report

During the duration of this program we made several advances in improving the understanding of the optical properties of ultrathin metal layers and their application in metamaterial metal-dielectric photonic bandgap filters (MDPBG) and other novel linear optical band-pass filters. The realization of high quality one dimensional metal-dielectric multilayer metamaterials requires an accurate knowledge of the refractive indices of the component materials. The transmittance through a MDPBG is limited by the absorption in the thin metallic layers throughout its structure. This absorption can be minimized if thin metallic layers, with thicknesses smaller than the metal's skin depth are used. However, the refractive index of thin metallic films has been shown to vary with film thickness and deposition systems, and even different "bulk" values have been reported. The refractive index is influenced in part by the surface roughness of the metallic films, which is in turn is affected by the adhesion between the metal layers and the adjacent dielectric layers. Through the surface roughness of the metal, non-radiative surface plasmon polaritons can be excited, leading to increased losses in such MDPBGs. Hence and important aspect that needs to be considered for the fabrication of such metal-dielectric meta-materials is to engineer the dispersion of such non-radiative surface plasmon polaritons in metallic structures with tuned optical properties.

We studied the effect of ultrathin (<1 nm) Titanium layers used as adhesion promoter for 20 nm-thick silver films. As substrate dielectrics 50 nm-thick layers of either SiO_2 or Al_2O_3 on glass were used. All layers were deposited by e-beam and the complex refractive index measured using a combination of ellipsometry, transmission, and reflection measurements. Through these experiments, it was found that: 1) the presence of Ti yielded optical properties closer to a 160 nm-thick silver layer; 2) The Ti thickness had a direct influence over the continuity and transparency of the silver films, yielding, for our deposition

conditions, the best results for Ti thicknesses of 0.5 nm; 3) that major discrepancies with respect to the optical properties of bulk silver were found in the imaginary part of the refractive index; and 4) that the dielectric surrounding has a small but noticeable impact over the refractive index of the metal layers. It is also worth noting that the properties of the silver layers were strongly dependent on the thickness of the Ti layer.

The permittivity of such thin silver films was also modeled from transmission and reflection data using an iterative numerical fitting routine based on the Newton-Raphson method in which the matrix transfer method was used to calculate the transmittance and reflectance of the structure. The starting point for the fitting routine was the measured bulk permittivity of Ag and, with each iteration, the permittivity was adjusted to minimize the discrepancy between the model and the measured data. This point by point fitting routine was repeated independently for each wavelength.

The frequency dependence of the permittivity in the visible region of the spectrum was described by the Drude model:

$$\varepsilon = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\gamma\omega}, \quad (1)$$

where ε_∞ is the sum of the interband contributions, ω_p is the bulk plasma frequency, and γ is a damping constant, which can be separated into a frequency-independent term γ_l and a frequency-dependent term β:

$$\gamma = \gamma_l + \beta\omega^2 . \quad (2)$$

Strictly speaking, ε_∞ is a function of frequency. Its dispersion could be calculated using the Kramers-Krönig relation from the interband absorption spectrum, however ε_∞ was approximated as a constant.

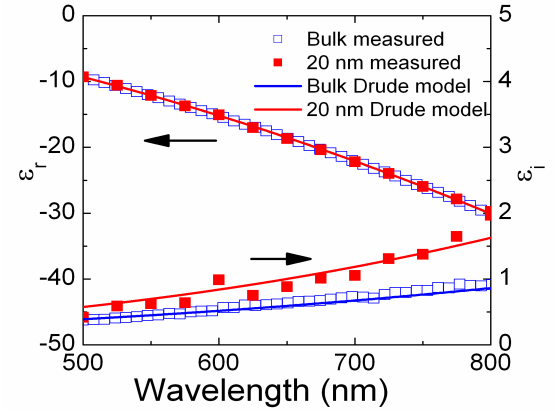

Figure 1. Measured and modeled complex permittivity of 160 nm and 20 nm silver layer.

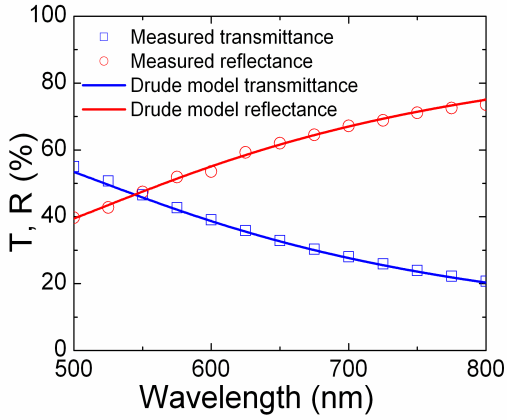

Figure 2. Measured and modeled optical properties of 20 nm Ag film.

The measured dispersion of the permittivity of the 160 nm-thick Ag film is shown in Fig. 1. The permittivity was fitted to the Drude model with $\varepsilon_\infty = 4$, $\omega_p = 1.38\times10^{16}$ s^{-1}, $\gamma_l = 2.73\times10^{13}$ s^{-1}, and $\beta = 5.9\times10^{-18}$ s^{-1}. These numbers compare favorably with values from the literature. In the literature, it has

been shown that in bulk Ag if the grain size is smaller than the intrinsic mean free path (~52 nm) then γ will increase linearly with the inverse of the grain diameter. This arises from the fact that grain boundaries interrupt the free flow of electrons, so the mean free path is limited by the diameter of the grains. The value of γ is very close to the damping value of 2.6×10^{13} s^{-1} that would be expected from a large, perfect crystal of Ag, indicating that the grain size is on the order of 50 nm or more.

The permittivity and thickness values of the thin were also calculated from transmittance and reflectance measurements shown in Fig. 2 by using the same fitting process as described above. The thickness was determined to be 19.7 nm and the permittivity was fitted to the Drude model with parameters identical to the bulk film except with $\gamma_1 = 8\times10^{13}$ s^{-1}. This difference is due either to increased surface scattering in the thinner film or a smaller grain size. The complex permittivity is shown in Fig. 1.

Accurate determination of the linear optical constants of these thin-silver films also enabled the description of the ultrafast thermally induced nonlinear optical changes that occur in thin silver films upon being exposed to an ultrafast optical pulse. This is an important realization which clarifies the optical origin of the nonlinear optical response of MDPBGs, which have also attracted attention for their large nonlinear optical properties but have nonetheless being analyzed in the context of third-order nonlinear optical processes in silver.

An additional aspect of concern in fabricating this thin metal layers was finding dielectrics that do not corrode the metallic layer, therefore assuring enviromental stability. In the past we showed that Ag/Al_2O_3 yielded MDPBGs that were environmentally stable for several months and up to 150 ^oC. However, in that case, no adhesion layers were used. Therefore, to study the stability of the Ti adhesion promotion layer, we fabricated Ag (20 nm)/Ti(0.5 nm)/SiO_2 films and subjected them to various extreme environmental conditions, including exposure to atmosphere for 7 days, heating to 200 ^oC in air for 1 hour followed by an ultrasonic bath in water for 20 minutes. As show in Figure 3, the transmittance of a 20 nm Ag film remained unchanged after all tests. This confirms that the combination of selected materials is environmentally stable and particularly that the presence of Ti does not promotes unwanted corrosion or oxidation of the silver layer.

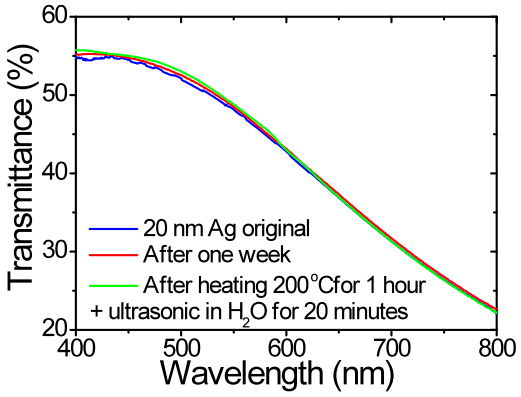

Figure 3. Complex refractive index of 20 nm silver layer sandwiched between 50 nm dielectric layers, with and without titanium adhesion layers

Having accurate models of the linear permittivity of silver and a combination of materials and processes that yielded environmentally stable metal-dielectric structures also allowed the demonstration of a structure similar to the MDPBG, namely the induced transmission filter (ITF). The ITF, originally described in 1957 by Berning PH and Turner AF, can be considered as a special class of meta-material which presents an interesting opportunity for the creation of narrowband linear and nonlinear optical filters because it contains only a single metal layer; as such, it is a simpler structure to fabricate and analyze than MDPBGs.

An ITF can be considered as a Fabry-Perot cavity where the Ag film is one mirror, albeit somewhat lossy, a ¼ wave stack is the other and a dielectric layer is the cavity spacer. In a high-finesse Fabry-Perot, where the total reflection at the peak wavelength is zero, two conditions must be met. First, the reflectances of the two mirrors must be equal; second, the round-trip phase within the cavity must be an integer multiple of 2π. In the ideal ITF, the first condition is met by choosing the number of periods

and the refractive indices of the ¼ wave stack such that its reflectance is equal to that of the Ag film. The second condition is met by choosing the thickness of the spacer layer to compensate for the non-zero phase shift of the reflection off the Ag film and the dielectric interface.

In order to demonstrate the properties of ITFs we fabricated two ITFs with the structure:

$$G\ (HL)_x\ R\ Ag\ P, \quad (3)$$

where G represents the glass substrate; H and L represent a ¼ wave layer of the high index and low index materials, in this case Ta_2O_5 and SiO_2, respectively; R represents a spacer layer of 75 nm SiO_2; Ag represents a 30 nm layer of Ag; and P represents a 410 nm protective coating of SiO_2. The parameter x defines the number of periods in the ¼ wave stack; for ITF 1, $x = 5$, and for ITF 2, $x = 4$. This is the only difference between the structures. The structures were designed to have center wavelengths at 600 nm, so using the refractive index values, measure by ellipsometry, of 2.03 for Ta_2O_5 and 1.46 for SiO_2, the ¼ wave thicknesses were 74 and 103 nm, respectively. The thickness of the protective layer was chosen to be equal to a full wavelength so that it does not affect the transmittance at the peak wavelength.

We simultaneously fabricated a structure that included only the 30 nm thick Ag layer and the protective coating. This reference film was deposited on 50 nm SiO_2 so that the dielectric environment surrounding the Ag film was identical with the ITFs.

Figure 4a shows the measured linear transmittance of all three structures, and Fig. 4b shows the measured transmittance, reflectance and absorptance of ITF 1 compared with simulations. As it can be seen from Fig. 4, an ITF greatly enhances the transmittance of single Ag film over a narrow bandwidth while preserving a high out of band rejection. Because ITFs maximize the potential transmittance of a given metal film, silver films as thick as 70 nm can in principle be used on ITF designs that achieve transmittances as large as 70 % while offering very high out of band rejection characteristics. However, the primary limitation to achieving such structures results from losses introduced primarily by the metal surface roughness.

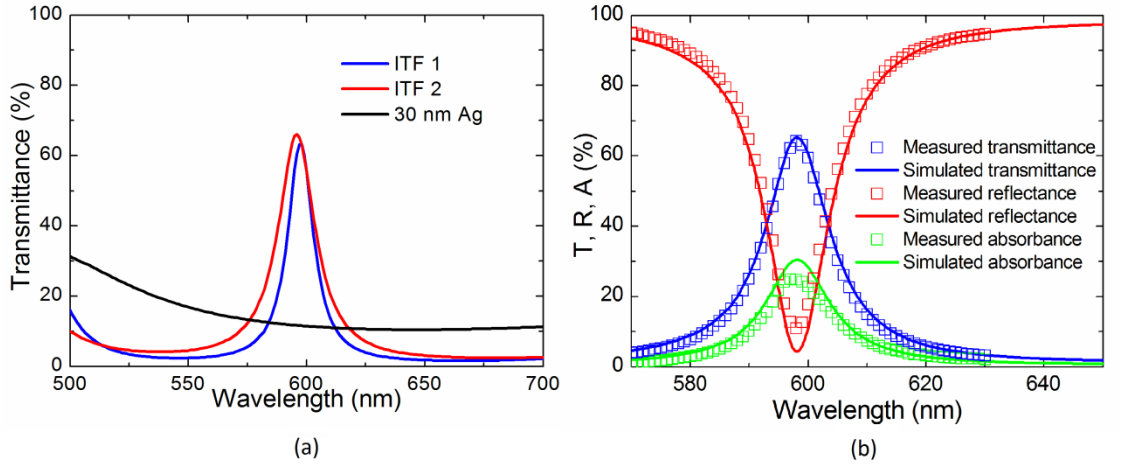

Figure 4. a) Measured transmittance of ITFs and a 30 nm-thick Ag layer; b) Comparison of measured and modeled transmittance, reflectance and absorptance of ITF1.

The surface roughness increases the optical losses of ultra-thin metallic films by creating the conditions for the efficient excitation of surface plasmon polaritons (SSP). The excitation of SPPs depends upon the dielectric permittivity of the metal but can be greatly assisted by the surface roughness of the films. Although the presence of transition metal inter-layers such as Ti decreases the surface roughness, the

low refractive index (or very negative real part of its dielectric constant) of Ag favors the excitation of SSPs compared with other metals such as silver. Figure 5 shows a comparison of the calculated dispersion characteristics of Ag and Au. Here it is clearly shown that the excitation of SPPs can occur at higher energies, across most of the visible spectra, on Ag compared to Au, where it can occur only beyond 550 nm. Although Ag, being the most reflective of all noble metals across the visible, theoretically offers the possibility of creating meta-materials with the larger transparency by creating a dielectric environment that suppresses its reflectivity, it also presents the problem of enabling efficient SPP coupling across the visible range. Hence, to control the spectral dispersion of the SPP losses in thin metallic layers we studied the optical properties of Ag/Au bi-layer films. The small difference in surface energy between Ag and Au, allows the growth of optically continuous ultrathin layers of one on top of the other. Fig. 6 shows a comparison of the measured and simulated transmittance of a 9 nm-thick Ag layer and a 9 nm-thick layer with a 1 nm-thick Au layer on top. In both cases, a 0.5 nm-thick Ti interlayer was used prior to the Ag deposition. The simulations were carried out assuming "bulk" refractive index values.

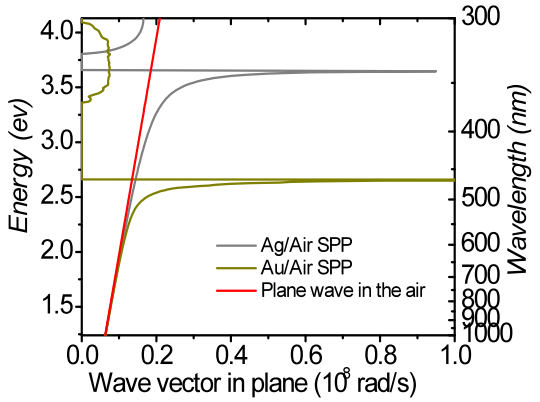

Figure 5. Comparison of SPP dispersion characteristics of Ag and Au.

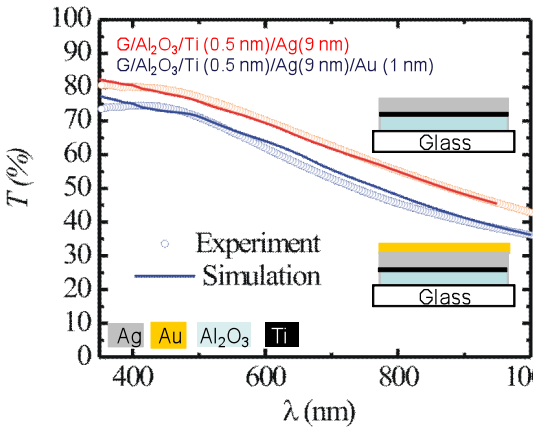

Figure 6. Measured and modeled complex permittivity of 160 nm and 20 nm silver layer.

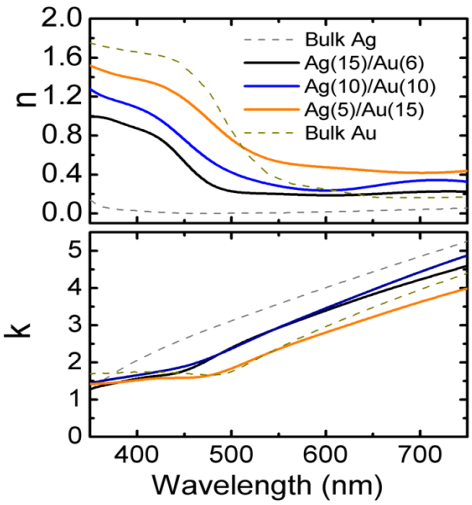

Figure 7. Complex refractive index values extracted by spectroscopic ellipsometry on Au, Ag and Au/Ag bi-layers.

Figure 7 shows a comparison of the refractive index of Ag/Au bi-layers with different thickness ratios and the bulk values for Au and Ag. The refractive index of the bi-layer metals were determined using spectroscopic ellipsometric measurements. This results show that, by controlling the thickness ratio, the effective refractive index of these bilayer metals can be tuned within the visible range. The ability to tune the refractive index, and consequently the dielectric permittivity, is an important degree of freedom which allows control over the spectral position of the SPP coupling losses while reducing the losses introduced

by interband transitions. These interband transitions severely limit the ability to create highly transparent meta-materials in the visible with noble metals such as Au or Cu.

Using a Ag(10 nm)/Au(10 nm) bi-layer ITFs structures were also fabricated. ITF structures are very sensitive to optical losses within the metal, and particularly SPP losses, because the higher finesse of this structure compared to MDPBGs, yields an increased optical field density within the metallic layer. Figure 8 shows the transmittance, reflectance and absorptance of such Ag/Au-based ITF with a peak transmittance of 83%, around 27% larger than the one obtained in a Ag-based ITF in the same spectral region. The thickness difference between the thicker silver layer and the thinner Ag/Au bi-layer explains only partially the big difference observed in peak transmittance between these structures; since in an ITF the peak transmittance does not scales linearly with metal thickness. Rather, the larger peak transmittance in observed in this bi-layer ITF structure is attributed to smaller SPP losses. If confirmed, this will make possible to fabricate meta-materials with narrow passband filter characteristics and with very high out-of-band rejection by increasing the thickness of the metal bi-layer.

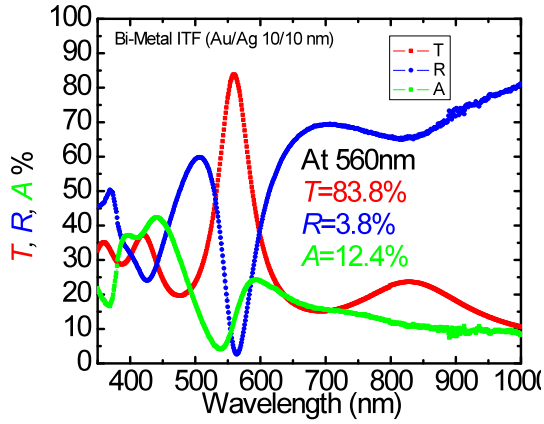

Figure 8. Transmittance of a Ag/Au (10/10 nm) induced transmission filter.

Finally, in addition to the previously mentioned metal-bi layers, thinner bi-layers were fabricated with the following targeted structure:

S1: Glass/Ta_2O_5(35 nm)/Ti(0.5 nm)/Ag(9 nm)/Au(6 nm)/Ti(0.5 nm)/Ta_2O_5(35 nm).

S2: Glass/Ta_2O_5(35 nm)/Ti(0.5 nm)/Ag(3 nm)/Au(3 nm) /Ag(3 nm)/Au(3 nm) /Ag(2 nm)/Ti(0.5 nm)/Ta_2O_5(35 nm).

The Ta_2O_5 layers at both ends of the multi-metal layer served has anti-reflection coatings which were expected to significantly increase the transmittance compared with the one expected from a bare multi-metal layer on glass. Furthermore, a Fabry-Perot resonator, the building block of MDPBGs based on a bi-layer metal structure used in a sample with geometry **S1** was also fabricated with the following geometry:

FP1: Glass/Ta_2O_5(35 nm)/Ti(0.2 nm)/Ag(9 nm)/Au(6 nm)/Ti(0.2 nm)/Ta_2O_5(70 nm)/Ti(0.2 nm)/Ag(9 nm)/Au(6 nm)/Ti(0.2 nm)/Ta_2O_5(35 nm).

Figure 9 shows the transmittance measured in samples **S1**, **S2** and **FP1**. The peak transmittance of samples **S1** and **S2** reaches a value of 76% at 568 and 528 nm, respectively. The slight shift in peak wavelength can be attributed mainly to differences in the Ag and Au morphology and distribution within the bilayer film. In sample **S1**, the first 9 nm-thick Ag layer has a sufficient mass-thickness to form a nearly continuous metal film. After the initial Ag layer, Au could be assumed to grow into a single continuous layer on top of Ag. On the other hand, in sample **S2** the initial deposition of Ag with a mass-thickness of 3 nm was non-continuous and formed nanometer-size metal islands upon which the following Au layer with a mass-thickness of 3 nm grew also in a discontinuous way. Hence, sample **S2** is expected to exhibit nanoscale domains of Ag and Au rather than be a one-dimensional smooth multilayer structure. These differences in the morphology and size of these Ag/Au domains are expected to lead to multi-metal layers with different effective refractive index and slightly different linear optical properties as shown in the figure. The transmittance in these structures was limited in part by reflectance losses due to a less than ideal admittance matching by the Ta_2O_5 layers and by linear absorption within the Ta_2O_5 layers, caused by the dissociation of Ta and O during the e-beam deposition.

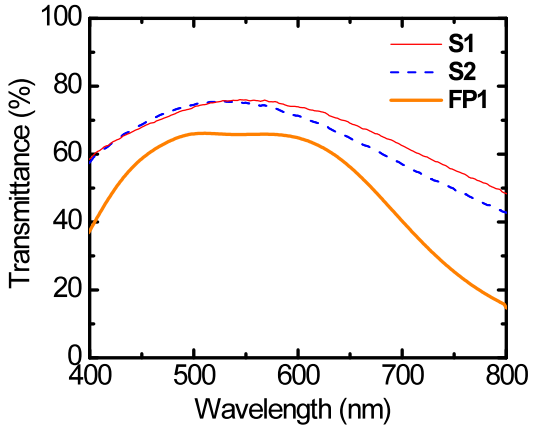

Figure 9. Transmittance measured on samples S1, S2 and FP1

List of Publications (Peer-reviewed and conference proceedings)

- D. T. Owens, C. Fuentes-Hernandez, J. M. Hales, J. W. Perry, and B. Kippelen, "*Nonlinear optical properties of induced transmission filters*," Optics Express **18** (18), 19101-19113 (2010).
- D. T. Owens, C. Fuentes-Hernandez, J. M. Hales, J. W. Perry, and B. Kippelen, "*A comprehensive analysis of the contributions to the nonlinear optical properties of thin Ag films*," Journal of Applied Physics **107** (12), 123114-123118 (2010).
- D.T. Owens, C. Fuentes-Hernandez, J. M. Hales, J. W. Perry, and B. Kippelen, "*A comprehensive study of the contributions to the nonlinear optical properties of thin Ag films*," in Active Photonic Materials III (SPIE Optics and Photonics, San Diego, CA) 7756 pp.77560K 2010.
- C. Fuentes-Hernandez, T. D. Owens, J. Hsu, R. A. Ernst, M. J. Hales, W. J. Perry, and B. Kippelen, in *The nonlinear optical response of transparent silver/gold multimetal layers*, SPIE proceedings, San Diego, CA, 2010, p. 77560G.

List of contributed and invited talks

- "*The Ultrafast Nonlinear Optical Properties of Induced Transmission Filters*," contributed talk at Conference on Lasers and Electro-Optics/Quantum Electronics and Laser Science Conference and Photonic Applications Systems Technologies, QFB5, Baltimore, MD, May 2011.
- "*A comprehensive study of the contributions to the nonlinear optical properties of thin Ag films*," contributed talk at SPIE Optics and Photonics, San Diego, CA, August 2010.
- "*The nonlinear optical response of transparent silver/gold multi-metal layers*," contributed talk at SPIE Optics and Photonics, San Diego, CA, August 2010.

- "*Metal-dielectric multilayer structures and their applications in nonlinear optics*" invited talk at the 14th International Conference on Laser Optics (LO-2010), St. Petersburg Russia, June 2010.
- *"Nonlinear optical properties of layered multi-metal nanostructures,"* contributed talk at Conference on Lasers and Electro-Optics/Quantum Electronics and Laser Science Conference and Photonic Applications Systems Technologies, San Diego, CA 2010.