Nonlinear Refraction and Absorption in Highly Transmissive One-Dimensional Metal-Organic Photonic Bandgap Structures


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Abstract: We report on the optical properties of a metal-organic photonic bandgap structure showing a peak transmission ~44 % and that enhances the nonlinear optical properties of bulk Copper by up to an order of magnitude.

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1. Introduction

One-dimensional metal-dielectric photonic bandgaps (MDPBGs) are attractive optical devices because they allow access to the extremely large and ultrafast nonlinear optical (NLO) response of metals [1,2], while keeping relatively large transmission windows in spectral regions where bulk metals are typically opaque [3]. The NLO response in bulk metals is dominated by absorptive processes in the visible spectra. Therefore, a good thermal compatibility between the layers forming the MDPBGs is desirable to prevent optical damage with high energy pulses. We have found that replacing the inorganic dielectric layers with polymeric layers, improves significantly the damage threshold and provides a material platform that can easily be tuned with the chemical flexibility brought about by the use of organic materials. In the present work, we report on the fabrication and characterization of the linear and nonlinear optical (NLO) properties of one-dimensional metal-organic photonic band gap structures (MOPBGs). Using the complex refractive index measured by spectral ellipsometry and the transfer-matrix method, we have designed MOPBG consisting of alternating layers of Cu, and a tetraphenyldiaminobiphenyl-based polymer (PTPD). The use of PTPD allows depositing continuous Cu layers of around 10 nm on top of the polymer layers, overcoming one of the persistent challenges for the fabrication of highly transmissive MOPBGs. The electrical continuity of the metal films was confirmed by conductivity measurements while spectral ellipsometric measurements were in good agreement with those expected for bulk Cu. Furthermore, using a transfer matrix method and the refractive index of bulk Cu, the linear transmission spectra was properly simulated when fitted with a Cu thickness of 10 nm.

Conceptually MOPBGs are not different from MDPBGs. In the linear regime, MOPBGs can be thought as stacked metal/organic/metal Fabry-Perot resonators with very low Finesse. Resonant transmission is achieved at \( \lambda_{\text{peak}} \) when the condition \( m \lambda_{\text{peak}} = 2 n_{\text{pol}} t_{\text{pol}} - \phi_1 - \phi_2 \) is met, where \( n_{\text{pol}} \) and \( t_{\text{pol}} \) are the refractive index and thickness of the polymer layer, while \( \phi_1 \) and \( \phi_2 \) are the phase accumulated by the backward and forward propagating fields inside each cavity upon multiple reflections throughout the structure. The MOPBGs presented, was designed to take advantage of the transparency window at the onset of the interband and intraband transitions on Cu. The thickness of the PTPD and Cu layers was selected to eliminate multiple resonances while preserving a large spectral bandwidth. The MOPBGs was fabricated with a combination of e-beam deposition and spin-coating techniques on glass substrates.

2. Results and Discussion

The MOPBG contains three resonant cavities and shows a peak transmission of around 44 % at 620 nm, five times larger than a solid Cu film with the same metal content, a spectral bandwidth of 150 nm (FWHM) with less than 5 % variation over the transmission window within a field-of view of at least 100 degrees, as shown in Figure 1a). The NLO properties of the MOPBG and two substructures were measured using closed and open aperture Z-scan
experiments, with 140 fs pulses, at 570 nm and 700 nm and its response compared with the complex nonlinear response of a continuous Cu film (~10 nm). We report measurements of the complex cubic NLO response of continuous Cu films as well as that of the MOPBGs.

In the single 10 nm thick Cu film, defining the refraction and absorption respectively as \( n(l) = n_0 + n_{2,\text{eff}} I \) and \( \alpha(l) = \alpha_0 + \beta_{\text{eff}} I \), we measured effective nonlinear refraction and absorption coefficients, \( n_{2,\text{eff}} = -2.4 \times 10^{-11} \text{ cm}^2/\text{W} \) and \( \beta_{\text{eff}} = 6 \times 10^{-6} \text{ cm/W} \) at 570 nm, while at 700 nm \( n_{2,\text{eff}} = 2.9 \times 10^{-11} \text{ cm}^2/\text{W} \) and \( \beta_{\text{eff}} = 1.7 \times 10^{-6} \text{ cm/W} \) were measured. The change in the sign of \( n_{2,\text{eff}} \) carries out in the spectral dependent NLO response of the MOPBGs. With no measurable contribution from the PTPD layers at these wavelength, we conclude that the NLO response is dominated by that of Cu. Figure 1b) shows the NLO response of the MOPBGs and substructures in the low irradiance regime where \( \Delta \phi = k n_{2,\text{eff}} L_{\text{eff}} I \) and \( q_0 = \beta_{\text{eff}} L_{\text{eff}} I \) are still good approximations to the measured irradiance dependence, normalized by the extrapolated NLO response expected of a continuous Cu thin film with a mass thickness equivalent to the total Cu content in the MOPBGs. In this regime, differences in the NLO response at the measured wavelengths correlate with the fact that a larger portion of the electric field is distributed inside the Cu layers at 700 nm than at 570 nm and with differences in sign of the nonlinearities in the metal at these wavelengths. Deviations from the expected linear irradiance dependence of \( \Delta \phi \) and \( q_0 \) were observed at either higher irradiances or as the number of resonant cavities was increased, providing further evidence of band detuning effects.

![Figure 1](CThI6.pdf)

Figure 1. (a) Experimental (symbols) and simulated (solid line) linear transmission of MOPBG structure and a continuous 49 nm Cu layer (dashed line). Inset shows the angular dependence and a photograph of the 1”x1” MOPBG; (b) Normalized nonlinear refraction (blue) and absorption (red) of the MOPBG and substructures.

3. Conclusions

Compared with typical semiconductors and organic materials, the presented MOPBG shows a remarkably large NLO response combined with a reasonably large transparency window, showing that it is possible to design and fabricate structures that maintain the very large nonlinear optical properties of metals and combine them with a large transmission in the visible spectral range.

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4. References