Highly reflective optical nanocavities for structural coloration by combining broadband absorber and Fabry–Pérot effects

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Abstract

Reflective structural colors are of interest for many applications as alternatives to dyes and pigments and many different solutions have been proposed. The ideal systems should provide high reflectance efficiency while keeping good chromaticity and offering tunability throughout the visible spectral range. It is challenging to achieve such combined features with a simple single structure. Here we address this challenge using a concept that combines the Fabry–Pérot effect with a broadband absorbing layer. Our easy-to-fabricate structures form highly reflective optical nanocavities with improved chromaticity compared with the two separate concepts. The addition of an additional cavity layer and a transparent top coating further improves the chromaticity and allows the formation of black surfaces.

Supplementary material for this article is available online

Keywords: structural colors, Fabry–Pérot, optical cavity, broadband absorber, reflective displays

(Some figures may appear in color only in the online journal)

1. Introduction

Many different approaches have been proposed for generation of reflective structural colors [1, 2], including plasmonic systems [3–9], Mie resonators [10–13], diffraction gratings [14], photonic crystals [15–17], and various types of optical nanocavities [1, 18–23]. Metal-insulator-metal (MIM) optical cavities based on the Fabry–Pérot (FP) effect are of interest owing to ease of fabrication and simple tunability of the resonance by changing the insulator thickness [24]. Compared with plasmon-based systems, MIM optical cavities are typically easier to fabricate and tune and they can provide high reflection efficiency. Nonetheless, while they can be efficient for transmissive color filters [25], it is less straightforward to use them for reflective structural coloration due to their spectrally sharp absorption and broadband non-resonant reflection. These issues have been addressed using porous or structured top metal layers [26], including plasmonic metal nanohole layers [27, 28]. Those strategies can be used to form a reflection peak by suppressing broadband reflection via material and nanostructure absorption. Another possibility to create cavities with reflection peaks is to replace the top mirror of a FP cavity with a broadband absorber [29, 30]. Such structure suppresses reflection at all wavelengths except in the range for which interference between incident and reflected light forms a minimum at the position of the absorbing layer. This principle has been exploited using phase-changing materials
for color tunability [31] and in MEMS-based color reflective displays [32]. These absorber cavities have the advantage of forming a proper reflection peak while maintaining high absolute reflection. However, they can suffer from poor chromaticity due to comparably large peak widths and reflection tails.

Here, we present hybrid planar optical nanocavities that produce reflective structural colors by combing the FP effect and a broadband absorber. These hybrid cavities are easy to fabricate, they provide good chromaticity combined with high reflection efficiencies, and the colors can be easily tuned throughout the entire visible spectrum and beyond.

We chose chromium as broadband absorber material because of its high and uniform imaginary permittivity in the whole visible range and its low real permittivity. It is also abundant and easy to deposit by standard techniques. An additional gold layer on top of the absorber improves chromaticity through the FP effect. This synergy effect takes advantage of the resonant FP absorption being red shifted from the position of maximum reflectance of the broadband absorber effect, thereby resulting in suppression of the reflection tail at longer wavelengths. This function resembles that of recently reported FP cavities with lossy top mirrors [33], but with the added possibility to separately tune the two different effects. To evaluate performance and improve understanding, we use experiments and simulations to compare the hybrid cavity (figure 1(c)) with the separate concepts of the FP cavity (figure 1(a)) and the broadband absorber cavity (figure 1(b)). We also present a double hybrid cavity that comprises an additional spacer layer and absorbing layer (see figure 1(d)). The hybrid cavity ensures efficient suppression of the red tail of the spectrum compared with the broadband absorber cavity. The double cavity further suppresses broadband reflection and improves chromaticity with only a small decrease in peak reflectance. Adding transparent dielectric top coatings further suppressed background reflection for all absorber-based cavities, resulting in significant improvements in chromaticity. In addition to full tunability in the visible spectrum, we demonstrate the possibility to form black surfaces by decreasing spacer thicknesses and tuning resonances into the ultraviolet.

2. Materials and methods

2.1. Fabrication

Optical cavities were fabricated using Corning® size 2 glass cover slides as substrates, cleaned by ultrasonication with a Mucasol® solution in deionized water, deionized water, acetone, IPA and then rinsed in IPA and dried with a nitrogen gun. The substrates were then cleaned by UV-ozone for 15 min. A 70 nm thick aluminum (Al) mirror was thermally evaporated onto the substrates at 20 Å s\(^{-1}\) with a Vaksis 2 system. This was followed by spin coating a 4% PMMA 996 K molecular weight solution in anisole for 60 s at various speeds to vary the thickness. Thicknesses were measured with a mechanical profilometer (Dektak) to obtain a spin-coating calibration curve. The top gold (Au) and/or chromium (Cr) layers were then thermally evaporated onto the PMMA to finalize the cavities. Black cavities required lower spacer thicknesses, for which we used 15 mg ml\(^{-1}\) PMMA MW 120 k in butanone, spin coated at 6000 rpm for 40 s. A 15 min baking step on a hotplate at 80 °C was added to improve film stability.

2.2. Simulations

Simulations were carried out with Comsol Multiphysics\(^{®}\) using the wave optics module in 2D. Periodic boundary conditions were used in the lateral direction of the cavity (using a periodicity of 300 nm) and we set the maximum mesh dimension to 40 nm. Incident light at normal incidence was defined as transverse-magnetic, but as expected, no meaningful differences were seen for transverse-electric light.

2.3. Measurements

Reflectance spectra were obtained using an optical microscope with a 50× objective (Nikon Eclipse L200N) connected by an optical fiber to a spectrophotometer (100 ms integration time averaged over five spectra (Ocean Optics, QePRO)). The reflectance data were normalized versus a silver mirror reference. For the black cavities we also measured the UV spectrum (200–400 nm) using a deuterium lamp (SL3, StellarNet Inc.) connected to a reflection probe (RP20, Thorlabs) at normal incidence in a probe stand (RPS-SMA, Thorlabs), still using the same spectrophotometer. As reference for these UV measurements, we used the same type of aluminum mirror as used for the cavities (because silver has a strong absorbance in this spectral region).

3. Results

Figure 1 presents the different types of planar optical cavities that we investigate and compare in terms of structural color generation. Each row corresponds to one type of cavity, with the first column (i) illustrating the basic cavity structures. The second column (ii) presents the corresponding optical near-field profiles (electric field norm) at maximum (\(R_{\text{max}}\)) and minimum (\(R_{\text{min}}\)) reflection. The third column (iii) shows simulated (dashed lines) and experimental (solid lines) reflectance spectra for example cavities, while the fourth column (iv) shows variation in the generated reflected colors for each type of cavity upon changing their spacer thickness. To illustrate the different physical mechanisms of the cavities, they were all fabricated and simulated with the same bottom mirror (70 nm aluminum) and the same spacer thickness (173 nm, as determined experimentally for PMMA films spin coated at 2500 rpm), except for the double cavity where the thickness was 184 nm (2200 rpm).

3.1. FP cavity

Figure 1(a) shows the traditional FP cavity, with a 20 nm thick layer of gold as semitransparent top mirror. The FP cavity effect results in a reflection dip at 642 nm (figure 1(a)(iii)), at which the optical field is strongly confined inside the
cavity (figure 1(a)(ii)). The experimental results are somewhat blue shifted compared with the simulations, as attributed to small differences in layer thicknesses and not perfectly flat gold when deposited on PMMA at low thicknesses. The first order FP resonance wavelength ($\lambda_{\text{FP}}$) can be determined by $\lambda_{\text{FP}} = 2(nL + L_{\text{Al}} + L_{\text{top}})$ [34–36], where $n$ and $l$ are the refractive index and thickness of the spacer layer, respectively. $L_{\text{Al}}$ and $L_{\text{top}}$ are the (wavelength dependent) contributions to the effective cavity length due to optical penetration into the aluminum and top gold mirror, respectively. Penetration of the optical field into the metals, and corresponding modifications to the phase shift upon reflection at the dielectric–metal interfaces, increases the effective thickness of the cavity and redshifts the resonance wavelength compared to a FP cavity made of perfect metal mirrors, for which the nodes would be at the dielectric–metal interfaces (also see field profile in figure 1(a)(ii)). For a spacer thickness of 173 nm with refractive index around 1.49, the simulated FP resonance dip was at 675 nm, which indicates an extension of the effective cavity thickness $L_{\text{Al}} + L_{\text{top}}$ by around 80 nm. The main disadvantage of FP cavities is that they produce absorption peaks instead of reflection peaks, which makes them less suitable for RGB structural colors in reflection mode. However, in our example we use gold as top layer, which is not a good reflector in the blue region (see complex and real permittivity in figure S1, supporting information (available online at stacks.iop.org/JOPT/23/015001/mmedia)). Instead, the gold acts as a partial absorber in the blue, which leads to a pseudo-reflection peak. At the peak position (540 nm) there is a node of the reflected standing wave (from the bottom mirror).
Figure 2. Experimental reflectance data for the different cavities with (black dashed lines) and without (green solid lines) top coating, all with spacer thickness of 159 nm. (a) FP cavity (10 nm gold top layer). (b) Broadband absorber cavity (10 nm chromium top layer). (c) Hybrid cavity (5 nm chromium and 7 nm gold top layer). (d) Double cavity (10 nm chromium as middle spacer, 5 nm chromium and 5 nm gold as top layer).

at the position of the top layer (figure 1(a)(ii)). Yet, also for this gold-based FP system, the reflective color gamut is limited upon tuning the spacer thickness, as illustrated in figure 1(a)(iv) for simulated reflection spectra converted to data points in the standard CIE diagram.

3.2. Absorber cavity

The same principle of avoiding absorption using interference nodes makes it possible to produce reflection peaks with cavities for which the top mirror has been replaced by a broadband absorber (figure 1(b)). For such absorber cavities, the top layer absorbs light and thereby suppresses reflection at all wavelengths except when a node is formed at its position (see figure 1(b)(ii)). At higher or lower wavelengths, the node is spatially shifted upwards or downwards, causing higher intensity in the absorbing layer and correspondingly lower reflection. The first order resonance of an absorber cavity can to a first approximation be estimated by \( \lambda_{\text{Abs}} = 2(nt + L_{Al}) \), which predicts resonances that are blue shifted from the FP cavity with same spacer layer. The reason for the blue shift is that the absorber resonance coincides with a node at the position of the top layer while the FP cavity effectively extends further and has considerable field strength in the top metal layer at resonance (compare the right part of figure 1(a)(ii) and the left part of figure 1(b)(ii)). The approximate absorber resonance condition indicates independence on top layer thickness, which agrees with simulated systems using (hypothetical) absorbers with same real refractive index as the spacer layer (and for fixed center position of the absorbing layer fixed, see figure S4). However, in real cavity systems, the top layer itself may contribute to modifications of the node position by modifying the refractive properties of the propagating wave. While several materials can be used as broadband absorbers, we chose chromium [30], because it has low magnitude real permittivity and high imaginary permittivity that is fairly constant throughout the visible wavelength range (see figure S1, supporting information). These properties makes a thin chromium layer functioning more as an absorber than a mirror, and it indeed does not show any significant contribution to FP effects. Moreover, chromium is an abundant metal that is easy to deposit with standard techniques. At an optimized chromium thickness of 10 nm, the fabricated absorber cavity produces a reflection peak at 552 nm, with an experimentally measured reflectance of 85%. As expected, the reflection peak of the absorber cavity is blue shifted from the resonance dip of the FP cavity. While the absorber cavity provides high brightness, the chromaticity is limited due to relatively large
peak width and high tails on both sides of the reflection peak. This can be seen from the CIE diagram (see figure 1(b)(iv)) for absorber cavities with different spacer thicknesses, with particular limitations in generating green colors.

3.3. Hybrid cavity

The fact that the reflection dip of the FP cavity (figure 1(a)(iii)) is red shifted from the reflection peak of the absorber cavity (figure 1(b)(ii)) means that the two effects can be combined. Indeed, this agrees with the pseudo-peak observed for the gold-based FP cavity (figure 1(a)) and which has also been reported for Ni-based cavities for which the metal simultaneously act as mirror and absorber [37]. Here, we introduce a double top layer that allows for the combination and independent optimization of the two effects (see figure 1(c)). In this hybrid cavity, the FP effect is controlled by a reflective metal while a broadband absorber ensures the reflective nature of the peak. Our cavity was optimized using a 7 nm thick gold layer on top of a 5 nm thick Cr layer, leading to a bright reflection peak with maximum experimentally determined reflection of 92% at 541 nm (figure 1(c)(iii)). While there is no significant change in peak position or width compared to the chromium-based absorber cavity (figure 1(b)(iii)), the added FP effect originating from the gold suppresses the reflectance tail towards the red region. This tail suppression is particularly pronounced for the experimental results. The FP contribution can be seen via higher field confinement inside the cavity at the minimum reflectance wavelength (765 nm) compared to the chromium-only cavity (figures 1(c)(ii) and (b)(ii), shown at a wavelength of 800 nm for the absorber cavity since it does not provide a reflection dip). Simulations predict no substantial optical difference if the chromium is placed before the gold or vice versa but experimentally the results better matched simulations when evaporating chromium first followed by gold. Thicker gold layers sharpened the suppressive FP effect, but with the disadvantage of increasing the reflectance at yet longer wavelengths (figure S3, supporting information). Increasing the thickness of the absorbing chromium layer instead led to higher tail reflection because the chromium becomes more reflective at higher thicknesses due to its metallic nature. Nonetheless, the possibility to independently tune the FP and absorber effects by choosing the top metal thicknesses forms an advantage of these hybrid cavities. Varying the spacer thickness enabled tuning of the color, with improved chromaticity and color gamut compared with the FP cavity and

![Figure 3](image-url)
partly also compared with the absorber cavity, in particular for the blue region (figure 1(c)(iv)). However, it was still challenging to obtain high quality green color due to the relatively wide reflection peak.

3.4. Double cavity

To lower the peak width, and thereby enhance chromaticity, we introduce a double cavity for which the hybrid cavity was combined with an additional absorber cavity (see figure 1(d)(i)). The two spacer layers were kept at the same thickness and separated by a 10 nm chromium layer. As top layer, we used 5 nm chromium and 5 nm gold. In this way, we form standing wave nodes in both chromium layers, as shown in figure 1(d)(ii). The double cavity produces a significantly sharper reflection peak compared with the single cavities, with only minor loss of reflectance efficiency at the peak position (78% at 574 nm). The experimental reflectance was limited compared with the simulated results, primarily due to inhomogeneities of the second spin-coated dielectric layer over chromium. The gold layer has a low effect on the overall spectrum and mostly ensures further suppression of the tail reflection at wavelengths to the red of the peak. Varying the spacer thickness demonstrates the possibility to tune the color through the visible at high color purity (figure 1(d)(iv)). The chromaticity is significantly improved in the green region compared with the other types of cavities. For thicker cavities with peak in the red wavelength region, a secondary peak appearing in the blue restricts the chromaticity from reaching further into the right most corner in the CIE diagram.

3.5. Effects of the dielectric top coating

To further reduce broadband background reflection, we investigate effects of adding an additional PMMA layer on top of the cavities. Similar strategies have previously been shown to improve the color saturation for optical cavities [30], by reducing interferential effects and corresponding reflection in unwanted parts of the spectrum. For the FP cavity (figure 2(a)), there was no observable improvement of the reflection dip.
by adding a top PMMA layer, but we observed an improved suppression of the reflection at lower wavelengths where the gold also acts as absorber. This latter observation resembles the results obtained for the three other types of cavities, which all showed significant tail suppression and corresponding improvements in chromaticity after coating with PMMA (figures 2(b)–(d)). Particularly pronounced improvement was found when the top dielectric layer was of the same thickness as the cavity spacer layer (figure S2, supporting information). The extra metal–dielectric and air–dielectric interfaces guarantee a better amplitude matching of the destructively interfering fields. This causes higher absorption of the out-of-node wavelengths of the spectrum and thereby more effective tail suppression for the coated cavities. The effect was most pronounced for the absorber cavity (figure 2(b)), but it provided important improvements also for the hybrid cavity (figure 2(c)) and the double cavity (figure 2(d)). Figure 3 shows how the reflectance for these three top-coated absorber-based cavities evolves with spacer thickness (120–240 nm), plotted as a color maps from simulations (top row). The resonances of these cavities increase approximately linearly with increasing spacer thickness, in accordance with the discussion in section 3.2. The CIE diagrams in figure 3(b) show what colors the simulated spectra translate to while also presenting corresponding experimental results and data for non-coated simulated cavities. It is clear that the top-coated cavities produced structural colors with improved chromaticity over the non-coated cavities. Tunability is provided throughout the visible, as demonstrated in figure 3(c) via photographs of top-coated hybrid cavities with different spacer thicknesses. We also note that the difference in chromaticity and color gamut between the absorber cavity and the hybrid cavity was less pronounced for the coated systems compared with the non-coated systems.

3.6. Black cavities

To reproduce an image, it is important to not only be able to print red, green and blue, but also be able to produce black pixels. Naturally, this is not possible for a FP cavity that produces a reflection dip. By contrast, one additional advantage of absorber cavities is the possibility to obtain black surfaces, in the means of a rather uniform high absorption in the entire visible range using lossy metals [33]. Simulations show that this can be achieved by tuning the first order reflection peak into the ultraviolet region by decreasing the spacer thickness (figure 4(a)). The effect worked well for the absorber cavity and the double cavity, while the hybrid cavity showed significant reflection at longer wavelengths due to the gold top layer (which was also thicker than for the double cavity). The strategy worked relatively well for uncoated cavities (figures 4(b) and (c)), but the addition of the PMMA top coating (with the same thickness as the spacers) resulted in significant further suppression of the reflected light throughout the visible. The optimal spacer thickness was found to be 60–70 nm for all three types of cavities. Figure 4(b) demonstrates the principle experimentally for an absorber cavity (with 79 ± 3 nm spacer thickness), showing reflectance suppressed to 4%–8% throughout the visible after top coating, and reflectance lower than 14% when uncoated. The insets in figures 4(b) and (c) verify that the effect is due to the reflection peak being tuned into the UV region (with peak positions of 285 nm for the coated broadband absorber cavity and 300 nm for the coated double cavity). A similar performance was obtained for the double cavity configuration (2%–8% visible reflectance when coated). The higher reflectance in experiments compared with simulations is primarily attributed to non-uniformity of the thin spin-coated PMMA layer and imperfections in the thin metal layers. Photographs of coated absorber and double cavities (figure 4(d)) confirms that it is possible to obtain black surfaces for the same structure as used to create the RGB colors, by only tuning the spacer thickness.

4. Conclusions

Combining FP and absorber effects into a hybrid cavity made it possible to produce reflective structural colors that cover the entire visible spectrum via the spacer thickness. We showed that the hybrid cavity improve chromaticity compared with the separate concepts by suppressing tail reflection, particularly on the red side of the reflection peak. The chromaticity could be further improved, in particular for green color production, by adding an additional absorber cavity layer to form a double cavity. Yet further suppressed broadband reflection and improved chromaticity was achieved by the addition of a transparent top coating, with best performance for thickness equal to the spacer thickness. The cavities were highly efficient in terms of strong reflection at the peak position and they could easily be tuned by changing the dielectric layer thickness. In addition to tunability within the entire visible wavelength range, the absorber and double cavity structures could also provide black surfaces, using lower spacer thicknesses that tuned the reflection peak into the ultraviolet. The simplicity of the structures makes them suitable for cheap large scale production, and the concept can also be modified with respect to materials used for spacers, mirrors and absorbers.

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