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Control of metal color using surface relief metamaterial nanostructuring

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Continuously metallic metamaterials can be frequency-selective, providing a means to engineer the color of uncoated metal surfaces.

Metamaterials are artificial electromagnetic media achieved by introducing structuring on scales smaller than the wavelength of incident electromagnetic radiation. As such, they have many unusual and useful properties. Whereas conventional materials derive their optical characteristics from their constituent atoms and molecules, metamaterials enable us to design our own 'meta-atoms', and to thereby access new optical phenomena, from invisibility and cloaking to negative refraction.[1] They open up new possibilities for advancing the control, guiding, and amplification of light, providing novel solutions for energy and light generation, imaging, lithography, data storage, telecoms, sensing, security, and defense.[2] Here, we show how a new class of continuously metallic metamaterials can bring frequency selective surface (FSS) functionality to the visible light domain. This provides a mechanism for controlling a metal's reflection spectrum and thus its perceived color, without recourse to chemical modification (e.g. anodization) or application of secondary coatings (such as paints or dielectric multilayers). The colors produced can, by design, be polarization dependent or independent and are largely insensitive to viewing angle.

Frequency-selective surfaces are a well-established method of filtering electromagnetic waves, particularly in the microwave and radio frequency bands where losses are minimal.[3] They are recognized as the foundation of the modern field of metamaterials.[2] Spectral selectivity is commonly achieved by manipulating the balance between reflected and transmitted waves. This can be done either through cascaded partially-transmitting boundaries (akin to distributed Bragg reflectors in optics) or by using periodic arrays of resonators, such as conducting elements in a dielectric matrix or apertures in a conducting screen. In our recent work, we demonstrated a novel class of FSS operating exclusively in the visible/near-IR spectral domain, where losses are important.[4] Under these circumstances, the plasmonic response of metals enables the modification of reflection spectra using the sub-wavelength patterning of metal surfaces without violation of continuity (i.e. without perforation). Absorption resonances can be engineered through the formation of arrays of inscribed ('intaglio') or raised ('bas-relief') nanoscale elements with a depth or height on the order of 100nm. The integrity of the metal surface is maintained by this patterning method, which is well suited to high-throughput fabrication via techniques such as nano-imprint. Frequency-selection of these structures is underpinned by plasmonic losses in the constituent elements of the patterns.

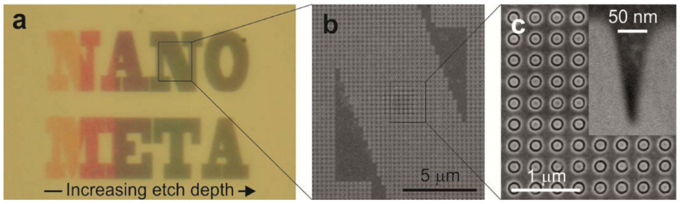

Figure 1. *Metallic structural color. (a) Optical microscope image of a gold surface, in which the words 'NANO META' are written with arrays of 170nm diameter rings. (b) and (c) Scanning electron microscope images of the rings, with characteristic slot cross-section shown inset of (c). Rings are milled to a depth varying (in six steps) from 60–200nm across the sample. (Figures modified from Zhang et al.[4])*

To demonstrate our technique, we inscribed a pure gold surface with an intaglio pattern of 170nm rings spelling out the words 'NANO META'; the result is vividly illustrated in the optical microscope image in Figure 1(a). We achieved different colors by simply varying the depth of the rings in the pattern, shown more clearly in the electron microscope images in parts (b) and (c) of the figure.

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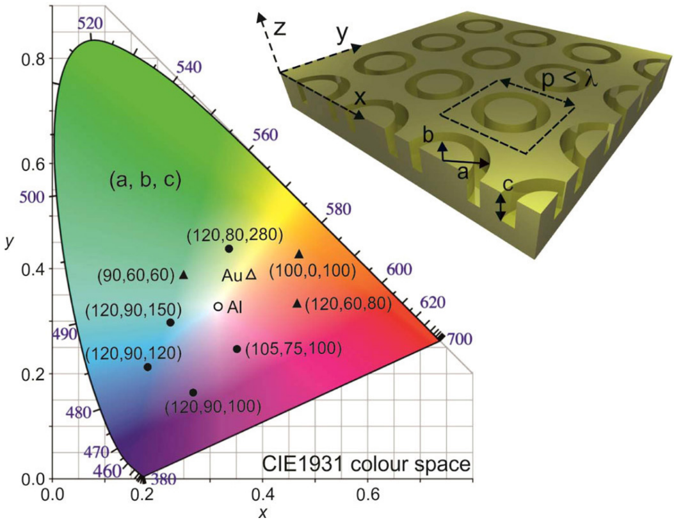

Figure 2. *Controlling the color of metals. This CIE1931 chromaticity diagram is overlaid with points corresponding to the simulated reflected colors of single-ring intaglio metamaterial patterns on aluminum (Al; circular symbols) and gold (Au; triangles). The values in brackets relate to the dimensions a, b and c illustrated in the inset graphic. (array period p=300nm in all cases). Points for the unstructured metals are denoted by open symbols. (Figures modified from Zhang et al.*[4]*)*

The functionality of intaglio/bas-relief designs, wherein subwavelength periodicity excludes diffraction effects, relies on the resonant plasmonic properties of the meta-atoms, which in turn are strong functions of geometry and size. The colors achieved by continuously metallic meta-surfaces are the result of selective enhancement of absorption in the visible spectral range, through the coupling of energy to plasmonic modes of the structures. The phenomenon is, therefore, best realized with good 'plasmonic' metals such as gold, silver, and aluminum.[5]

To determine how structural geometry affects the color of these continuously metallic metamaterials, we performed a finite element numerical analysis of single-ring meta-molecule arrays. Variations on this simple design, illustrated schematically in the upper right portion of Figure 2, can provide access to a significant color range on aluminum and gold surfaces, as shown in the lower left portion of the figure. The available parameter space for meta-molecule design and distribution is almost unlimited, although the technique, being based on the manipulation of absorption, is not suited to achieving the pure colors found at the boundaries of the color space. It does, however, have the advantage of generating colors that are independent of viewing angle (or at least no more dependent on angle than for unstructured metal surfaces), and it provides for the possibility of introducing anisotropy (polarization-dependent color) through structural asymmetry.

In summary, Intaglio/bas-relief photonic metamaterials offer a robust, flexible, and scalable paradigm for engineering the spectral response of metals in the visible domain. Potential applications range from the purely decorative (e.g. jewelry) to optical sensing and security (anti-counterfeiting features must be difficult to imitate without substantial advance investment in design and fabrication technology). We are currently working on the refinement of pattern geometries and fabrication techniques. Indeed, while proof-of-principle demonstrations have so far relied on focused ion beam milling and electron beam lithography, continuously metallic intaglio and bas-relief metamaterials offer a highly controllable mechanism for engineering and manipulating diffraction-free, angle-invariant absorption resonances. They may ultimately be implemented using simple, low-cost, scalable fabrication techniques (e.g. nano-imprint, template stripping), even on non-planar surfaces.

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