Second harmonic generation due to mesoscopic structuring in dielectric metamaterials

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Abstract: We demonstrate second harmonic generation by amorphous material after breaking its inversion symmetry by structuring: A resonant silicon metamaterial on an optical fibre tip converts infrared into red light that is guided along the fibre.

Second harmonic generation occurs in crystalline materials that lack inversion symmetry on the atomic scale, and at interfaces where inversion symmetry is broken by the material boundary. Here we demonstrate metamaterial with second order nonlinearity consisting of amorphous media without intrinsic second order nonlinear response. We demonstrate second harmonic generation due to breaking the inversion symmetry of a silicon film by mesoscopic structuring. Our approach enables metamaterial engineering of spectral position and magnitude of second order nonlinear optical effects.

The all-dielectric metamaterial consists of an array of chevron groove pairs milled into the core of a silica fibre and subsequently coated with a 90-nm-thick amorphous silicon layer. The unit cell size is $1.1 \times 1.0 \, \mu\text{m}^2$ and the overall dimensions are $22 \times 21 \, \mu\text{m}^2$, see Fig. 1(a). Due to a size difference of the chevrons contained in each unit cell, the metamaterial exhibits an asymmetric resonance with a quality factor of about 30 at 1500 nm wavelength (Fig. 1b), when illuminated with light polarized along the chevron symmetry axis (y-axis in Fig 1a).

To characterize the (second order) nonlinear response of the metamaterial it was illuminated by y-polarized, 200 fs optical pulses with 32 mW average power and 80 MHz repetition rate. The centre wavelength of the pulses was tuneable over the entire NIR range with a pulse bandwidth of about 5 nm. The light was focused onto the fibre-tip reaching a peak intensity of about 8 GW/cm². The radiation coupled into the fibre was then filtered to remove the pump as well as any third harmonic signal, and then detected using a photomultiplier tube. We observe second harmonic generation at the metamaterial resonance, see Fig. 1(b). Second harmonic generation did not exceed the noise level in control experiments on unstructured reference fibres with and without the silicon film, confirming that the observed second harmonic generation is caused by the metamaterial nanostructure. Accounting for the detection efficiency of the photomultiplier tube, we observe a second harmonic power of 60 fW and a power conversion efficiency of at least $10^{-12}$ in this proof-of-principle demonstration. We expect dramatic increases of second harmonic generation efficiency to be achievable with optimized metamaterial geometry.

In summary, our results show that second harmonic generation can be caused and engineered by breaking inversion symmetry with mesoscopic structuring. We observe second harmonic generation on a low-loss amorphous dielectric film and its resonant enhancement when the film is structured in a way that lacks inversion symmetry.

Fig. 1 Second harmonic generation caused by mesoscopic structuring. (a) Dielectric metamaterial lacking inversion symmetry. The structure covers the core of a cleaved optical fibre and consists of pairs of chevron grooves coated with a silicon layer of 90 nm thickness. (To prevent charging, SEM imaging took place with an additional gold coating that was subsequently removed.) (b) Detected second harmonic generation rate of metamaterial (red) and reference samples (blue) alongside the metamaterial’s linear transmission (black) as a function of the wavelength of the y-polarized pump beam.