Design of Metallic Nanostructures for Enhanced Nonlinear Optical Response

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Abstract: Four-wave-mixing from rectangular metallic nanocavities in a thin gold film is observed experimentally and discussed theoretically, and the cavity shape is optimized to provide enhancement of more than an order of magnitude.

Plasmonic devices can provide extreme localization of light and subsequent near-field enhancement, which can be used to boost nonlinear phenomena[1]. The strong intrinsic nonlinear response of noble metals makes plasmonic nano-structures the ideal candidates for the design of ultrasmall and/or ultrathin devices. In particular, thin metal films in which periodic nanocavities are fabricated give rise to strong extraordinary optical transmission and strong nonlinear response. Optimization of the geometrical parameters is shown to lead to controlled enhancement of the Four-Wave Mixing (FWM) response.

Two near-infrared femtosecond laser pulses (ω_1 = 800 nm and ω_2 = 1050 nm) are mixed in arrays of periodic nano-rectangles drilled in a thin gold film (Fig. 1a), leading to phase matched coherent emission at $\omega_3 = 2\omega_1 - \omega_2$ = 645 nm (Fig 1b). The linear transmission spectrum of each array is calculated and measured, and used for optimization of the FWM response. In turn, the FWM intensity dependence on the aspect ratio (AR) of the nano rectangles is measured, and calculated using 3D nonlinear finite differences time domain (3D-NL-FDTD). In this calculation, the field update equations are modified to include the nonlinear polarization induced in the material. The FWM response is further analyzed in terms of the coupled-mode theory (CMT), which assumes that the longitudinal modes propagate inside the cavities and exchange energy. The theoretical analysis can provide strategies for the rational design of nonlinear meta-materials.

Figure 1c depicts selected transmission spectra measured for different aspect ratios, showing resonance response at different frequencies. In figure 1d we show the dependence of the generated FWM intensity on the AR, demonstrating a 10-fold enhancement of the intensity of the generated FWM for the properly designed nanocavity. Figure 1e depicts the calculated FWM intensity, capturing the same strong dependence on AR, and lastly, figure 1f shows the field distribution for resonant (AR=2.1) and nonresonant (AR=1.0) cavities, clearly showing the enhanced field intensity for the resonant structure.

In conclusion, the observation of FWM from nanocavities is demonstrated experimentally and analyzed theoretically in terms of resonant modes within the nanostructures, leading to further options for designing of metamaterials for enhanced nonlinear optical response.

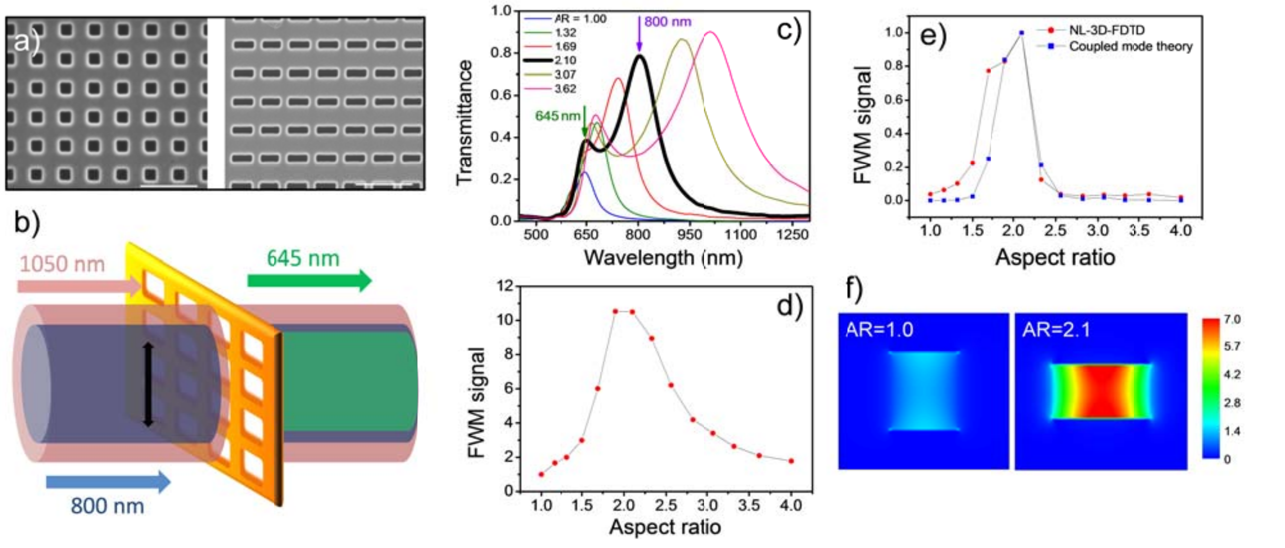

Fig 1. (a) SEM of the nanorectangles arrays. (b) Schematics of the FWM experiment. (c) Experimental transmittance of some arrays. (d) FWM intensity (normalized to AR=1.0) as a function of the AR. (e) NL-3D-FDTD calculations and coupled mode theory of FWM generation. (f) Field distribution at ω_1 = 800 nm in cavities with AR=1.0 and AR=2.1.

[1] M. Kauranen, A. V. Zayats, "Nonlinear Plasmonics", Nature Photon. **6**, 737 (2012).