Local optimization of nonlinear luminescence in disordered gold metasurfaces by far-field wavefront control

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Engineering the wavefront of light in random media allows the control of wave propagation in space and time by exploiting the spatial and spectral degrees of freedom introduced by multiple scattering.¹ To apply this far-field control strategy and focus electromagnetic energy at the nanoscale, it is necessary to introduce scatterers that feature strongly enhanced and confined optical fields such as plasmonic nanoantennas. In particular, semi-continuous gold films close to the percolation threshold feature high local field enhancements² but also propagating surface plasmon waves that can be controlled using a spatial light modulator.³

In this presentation, we demonstrate how controlling the phase of an incoming femtosecond pulsed laser on a chosen area of a disordered plasmonic metasurface allows us to optimize the two-photon luminescence (TPL) of gold at a given position of the sample. In practice, the incoming wave is shaped by a spatial light modulator (SLM), which is conjugated to the sample surface (see Figure 1-a). This provides us with a full control of the phase of the EM wave on the metasurface while maintaining a uniform illumination. Figure 1-b is a typical example of a widefield TPL image when exciting a plasmonic surface (gold filling fraction of 0.6) with a random phase pattern. The TPL signal originating from the center of the 35 µm x 35 µm phase pattern is optimized using a random iterative process as demonstrated in Figure 1-c.

The optimized TPL intensities are increased by an average factor of 50 for disordered gold metasurfaces that are close to percolation (see Figure 1d). When the filling fraction of gold is far from percolation, the enhancement factors decrease dramatically, demonstrating that the morphology and level of disorder of the plasmonic surface play an essential role in the wavefront control of nonlinear luminescence. Furthermore, we show that TPL intensities can be enhanced at any position of a percolated film. Since the TPL intensities are associated with strong local field enhancements, these results open exciting perspectives for the wavefront engineering of plasmonic hot-spots in disordered gold metasurfaces.

Figure 1. (a) Excitation and imaging setup. The gold metasurface (scale bar = 100 nm) is illuminated by a wavefront controlled femtosecond pulsed laser while the TPL signal is observed in full field with an EMCCD camera. Typical TPL image of the excited plasmonic metasurface before (b) and after (c) iterative optimization (scale bar = 2 µm). (d) Average TPL enhancement factor as a function of the gold filling fraction of the plasmonic metasurface.

References

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