Coherent Control of Nanoparticles' Nonlinear Response

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In our research, we utilize a sub-10 femtoseconds pulse shaper to study and control the Four-wave mixing (FWM) nonlinear response of LSPR excitations within the coherent regime. We use a spatial light modulator (SLM) to tailor the spectral phase of the pulse, in the attempt to complement the inherent LSPRs' spectral phase. Our results outperform the transform limited case by an 80% enhancement factor, confirming the coherent dynamics predicted by the anharmonic oscillator model. Our experimental method enables extraordinary control capabilities, which may be applied to arbitrarily shaped nanoparticles, bypassing symmetry-related restrictions which limited similar previous studies.

Experimental Apparatus

In our experiment we use a spatial light modulator (SLM) situated in the Fourier plane of a 4f system to shape the phase of an ultra-fast $7fs$ laser (Venteon, 650-1100nm). The use of an ultra-broadband laser enables us to explore the dynamics of resonant excitations with very short coherence timescales, such as localized surface plasmon resonances (LSPRs). Coherent control of LSPRs was recently demonstrated using the sum frequency generation (SFG) from Au split ring resonators (SRRs) [1]. This method, however, is limited to specific material geometries.

Here we present coherent control of LSPRs while monitoring the near degenerate four wave mixing (FWM) signal of Au nano-bars. As opposed to SFG process, this nonlinear process doesn't place geometrical constraints on the geometry of the sample under investigation. This enables us to study any type of geometry.

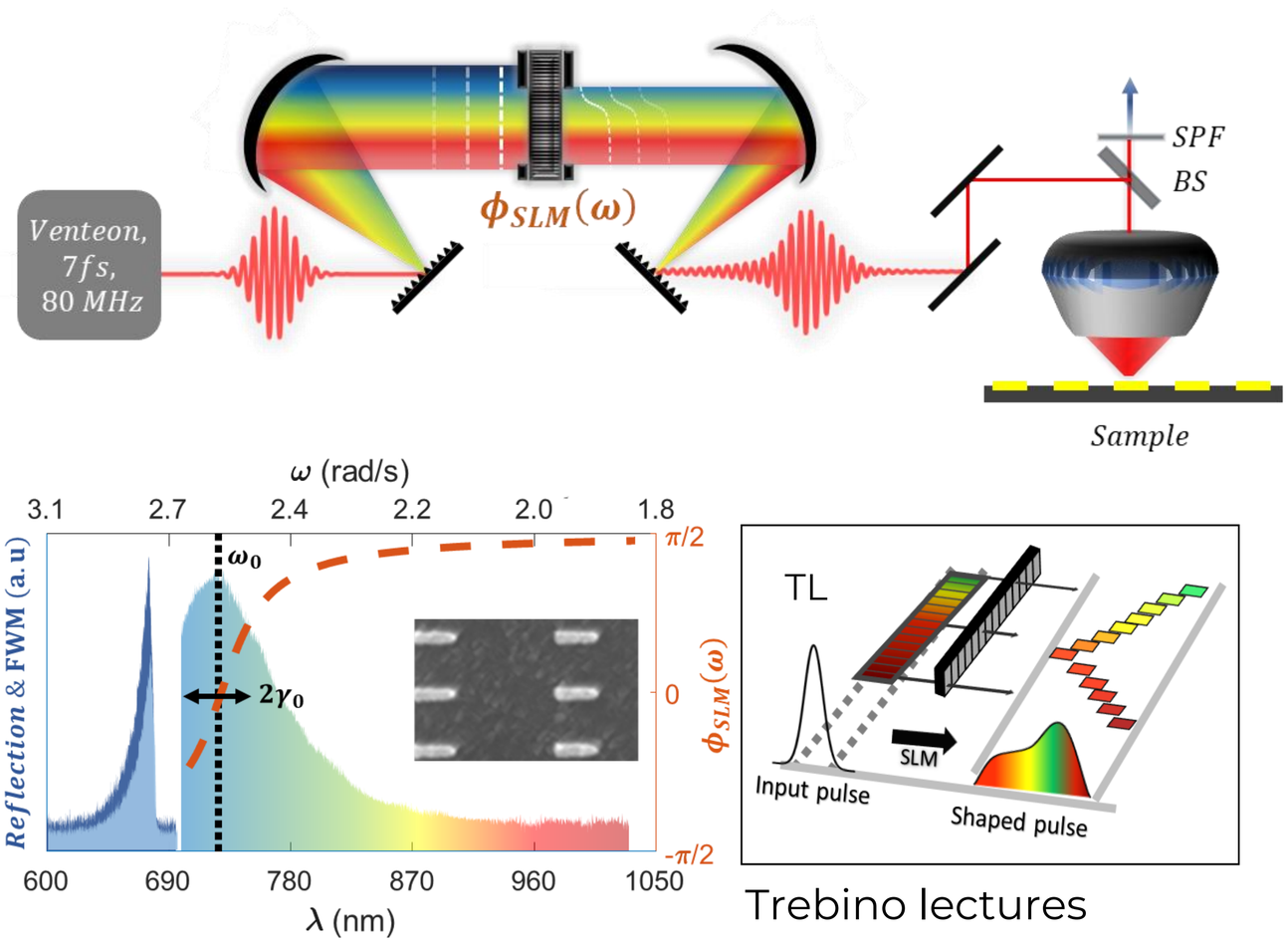

Figure 1 – experimental set up

The Model

We model the nanoparticles' response to the pulse as an anharmonic oscillator as depicted in the following equation:

$$\ddot{x} + 2\gamma_0\dot{x} + \omega_0^2 x = -\frac{eE(t)}{m} - \alpha x^3$$

the linear solution is:

$$\widetilde{x_0} = \frac{e}{m}\left|\frac{\tilde{E}(\omega)}{D(\omega)}\right| e^{i(\phi_E - \phi_D)}, while\ D(\omega) = \omega^2 - \omega_0^2 - 2i\omega\gamma_0$$

We calculate the third-order nonlinear response by using perturbation analysis. The FWM response is approximated by:

$$\widetilde{\delta x_0}(\omega) \propto -\alpha\frac{\widetilde{x_0}(\omega) * \widetilde{x_0}(\omega) * \widetilde{x_0}(\omega)}{D(\omega)}$$

To maximize this term, we demand that $\widetilde{x_0} = |\widetilde{x_0}|$. This can be achieved by applying the following spectral phase to our ultra-broadband pulse:

$$\phi_E = \phi_D = -\tan^{-1}\frac{2\gamma_0}{\omega_0^2 - \omega^2} \approx -\tan^{-1}\frac{\omega - \omega_0}{\gamma_0}$$

Results

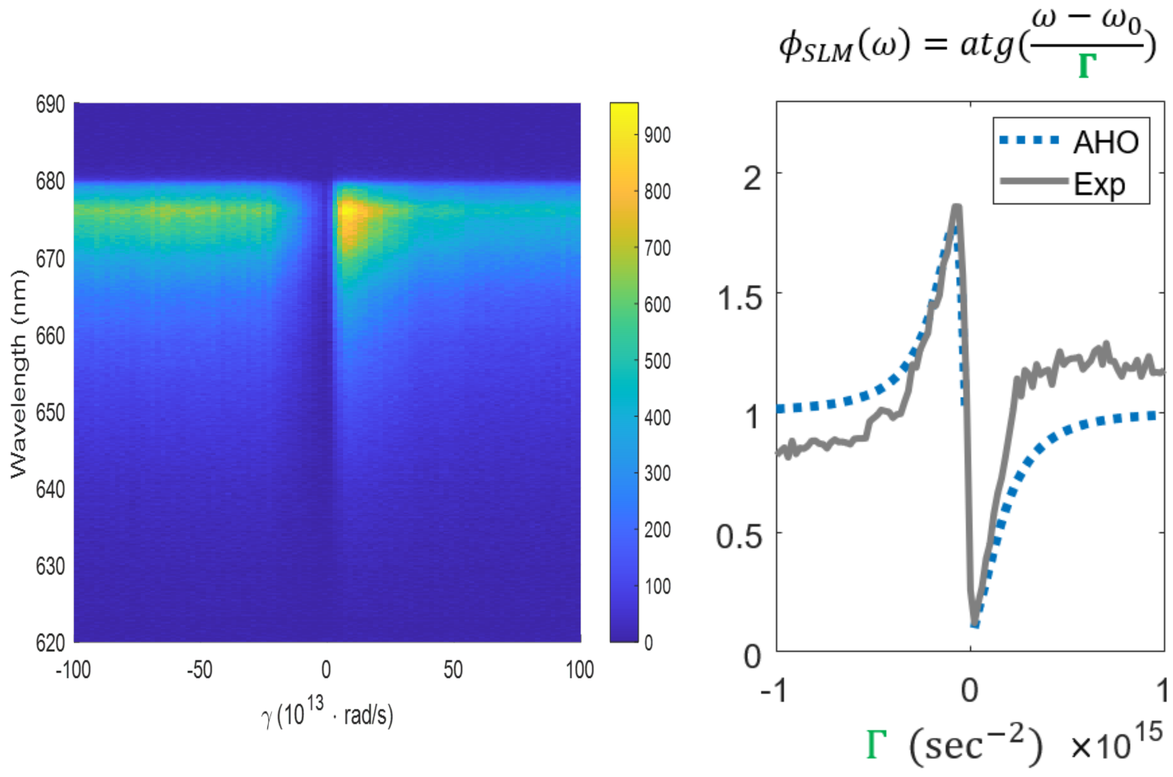

Figure 2 – experimental and theoretical results

Non-Instantaneous Phase Response

A transform limited (TL) pulse is a pulse where all frequency components interfere constructively, producing the largest peak intensity. It is commonly believed that the interaction of a transform-limited pulse with a nonlinear mediator, generates the greatest nonlinear optical response. This statement applies to non-resonant materials with an instantaneous optical response.

In resonant materials, however, the photo-excited carriers induce a non-instantaneous response [2]. This phenomenon gives rise to a non-trivial spectral phase inherent to the material. It has been shown that in this case, the TL pulse is not optimal [1].

To validate the model, we scanned the linewidth of an arctangent phase while holding ω_0 at a fixed value. In figure 2 we compare the FWM intensity as calculated in our model and as measured experimentally.

When the linewidth of the arctangent phase approaches the negative value of the LSPR's linewidth, complementing the inherent phase of the resonance, we observe an enhancement of 80% in the FWM signal relative to the TL case. It's worth noting that even though the peak intensity of the pulse is reduced, the material nonlinear response increases.

By applying the same arctangent phase, with the opposite sign we manage, with the same power distribution – but reversed in time, to show destructive interference of the FWM signal.

[1] Bahar, Eyal, et al. "Unlocking coherent control of ultrafast plasmonic interaction." Laser & Photonics Reviews 16.7 (2022): 2100467.

[2] Dudovich, Nirit, et al. "Transform-limited pulses are not optimal for resonant multiphoton transitions." Physical Review Letters 86.1 (2001): 47.