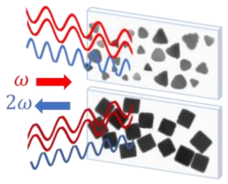

Characterization of Second-Harmonic Generation in Silver Nanoparticles for Spontaneous Parametric Down-Conversion

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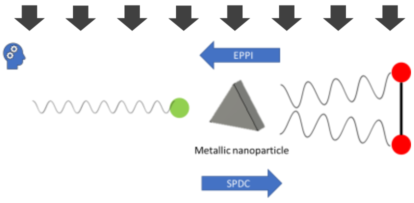
INTRODUCTION

- Broadband energy-time entangled-photon pairs (EPPs) are essential for advanced quantum technologies, such as spectroscopy[1], reaching beyond classical limitations.
- They are commonly generated through spontaneous parametric down-conversion (SPDC) in nonlinear crystal, where their bandwidth is limited by phase-matching requirements.

$$\varphi_{EPP} \propto sinc^2(L\Delta\boldsymbol{k}/2)$$

$$\Delta\boldsymbol{k} \equiv \boldsymbol{k}_p - \boldsymbol{k}_s - \boldsymbol{k}_i \qquad L - interaction\ length$$

- Metallic nanoparticles (NPs) are known for their exceptional capability of light-matter coupling at their localized surface-plasmon resonance (LSPR).
- Strong second-harmonic generation (SHG) by EPP interaction (EPPI) with NPs was suggested[2].



- Optimizing NPs for detectable non-phase matched SPDC ($L \ll \lambda$) requires classical-light SHG characterization measurements.

METHOD

- Properties of EPPI or SPDC can be deduced from classical-light SHG measurements performed using our reference-free hyper-Rayleigh scattering (RFHRS) method[2], approximating the NPs as Hertzian dipoles at the second-harmonic frequency.
- This method yields:

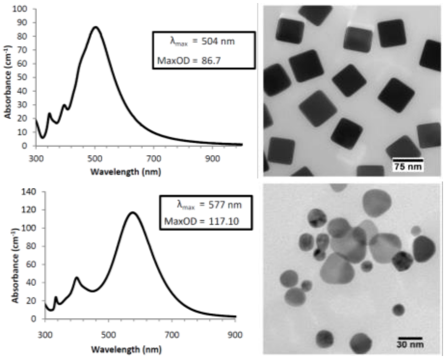
$$I_{2\omega} \approx \frac{2V\omega^4}{\pi^2\varepsilon_0^3 c^5 r^2}\langle C_m\beta_m^2 + C_{NP}\beta_{NP}^2\rangle I_\omega^2 T_{SH}$$

$$\eta\sigma_c = \frac{8\hbar\omega^5\beta_{NP}^2}{3\pi\varepsilon_0^3 c^5}$$

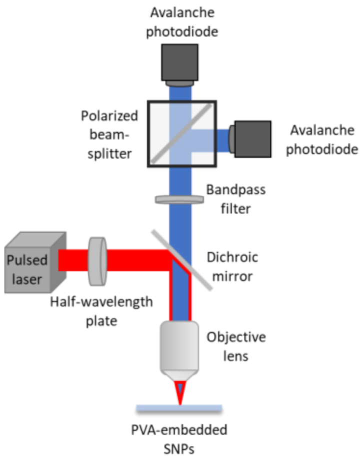
- Here, V is the interaction volume, r the distance of the collecting lens from the sample, C_m (β_m) and C_{NP} (β_{NP}) are concentrations (hyperpolarizabilities) of the medium and the NPs, T_{SH} is the transmission of the second-harmonic signal through the system, η is the quantum yield of the SHG process, and σ_c is the classical two-photon interaction cross-section.

SAMPLES

Silver nanocubes (SNCs) and nanotriangles (SNTs). Samples were diluted and then embedded in polyvinyl-alcohol (PVA) on a glass slide.



OPTICAL SETUP



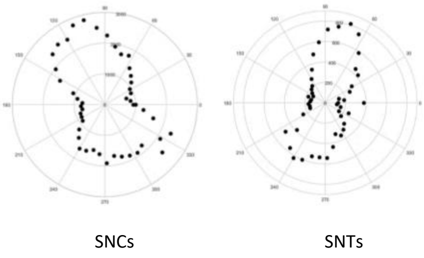
For SNCs we used an incident wavelength of $980[nm]$ corresponding to SH wavelength at their LSPR peak, while for SNTs we used $940[nm]$ which corresponds to SH wavelength at a local minimum of their spectrum.

RESULTS

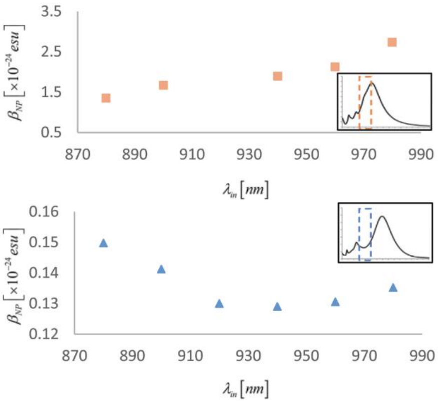
Hyperpolarizability and two-photon interaction cross-section[3]:

Sample	$\beta[\times 10^{-25} esu]$	$\eta\sigma_c[GM]$
SNCs	16.45	520.96
SNTs	1.89	8.5

Polarization:



Hyperpolarizability dependence on excitation wavelength:



CONCLUSIONS

- NPs' hyperpolarizability is in excellent agreement with theory[4] and previous reports[5], a validity proof of the RFHRS method.
- NPs' effective nonlinear coefficient (d_{NL}) is much smaller than crystals like BBO, but NPs are highly tunable and the bandwidth of the EPPs is not restricted by phase-matching.
- Dipolar emission pattern, polarization of EPPs will be dictated by largest $\chi^{(2)}$ component.
- Hyperpolarizability shows clear dependence on NPs' spectral features, thus tuning of LSPR wavelength is critical for SPDC enhancement.
- Linear dependence of the SH signal on NPs' concentration, suggesting incoherent process.

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