Modulating the Temporal Dynamics of Nonlinear Ultrafast Plasmon Resonances

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Nonlinear plasmon modes (NLPs) supported by metallic nanoparticles play a vital role in enhanced nonlinear optics, high-resolution optical imaging and advanced spectroscopy applications [1]. However, the ultrashort lifetime of these intense nonlinear fields inhibits their effective use in the vast applications of quantum plasmonics. These NLPs formulate through the spatial convolution of fundamental plasmon modes (FM) and decay shortly within 10 fs [2]. FM, being a constituent part, contributes to the intensity and spatiotemporal dynamics of nonlinear plasmonic signal. To acquire pure nonlinear effects, its spectral and temporal independence from the FM is of significant importance. In this paper, we demonstrate the lifetime enhancement of second harmonic (SH) plasmon mode, supported by Au nanoparticle (AuNP), through the resonant coupling with long-lived quantum emitter (QE) (Fig.1 (a) inset) [3]. We utilize coupled harmonic oscillator model and the total Hamiltonian of the system given in Eq.1 consists of eigen energies of plasmon modes (\(\alpha_j\)) and QE, interaction term with coupling strength (\(\kappa\)), interaction of incident light (\(\omega\)) and the convolution of FM along with nonlinear susceptibility \(\chi^{(2)}\).

\[
\hat{H} = \sum_{j=1}^{2} \omega_j \hat{a}_j^{\dagger} \hat{a}_j + \omega_{eg} |e\rangle \langle e| + \kappa |g\rangle \langle g| + \epsilon_1 \hat{a}_1^{\dagger} e^{-i\omega t} + i\chi^{(2)} (\hat{a}_1^{\dagger} \hat{a}_1 + \hat{a}_2^{\dagger} \hat{a}_2) \tag{1}
\]

The complex amplitude of the SH mode is derived from the Heisenberg equation of motion, \(\partial_t \hat{a}_j = i[\hat{H}, \hat{a}_j]\) and the lifetime of SH mode is evaluated as time-weighted over the mean value of plasmon intensity as \(\tau = \frac{\int_0^\infty \int_0^\infty E_{266}^2 E_{265}^2 \ d\omega d\omega \tau_{eg}}{\int_0^\infty \int_0^\infty E_{266}^2 E_{265}^2 \ d\omega d\omega} \) [4]. For \(\gamma_{eg} = 10^{-6} \omega\), 800 times enhancement is obtained in the lifetime of SH field, red dotted curve (Fig.1 (a)).

For a more realistic visualization, we performed 3D numerical simulations to compute the field-time response of the AuNP-QE system. We evaluated the response of SH modes in the absence and presence of QE through the finite difference time domain (FDTD) method. In the absence of QE, the power spectrum (red line) exhibits SH intensity which decays down shortly in 10 fs. Whereas in the resonant interaction of QE with SH field at (\(\omega_{266} \approx \omega_{eg}\)), a three-fold enhancement in the hotspot intensity of SH mode is observed. Moreover, due to the small decay rate of QE, the SH field oscillates for longer with an average lifetime \(\tau\) of 129 fs (Fig.1 (b)) after the natural decay time of FM mode. Our approach show coherent control over spatial and temporal dynamics of SH plasmon modes purely which pave its way for designing efficient on-chip nonlinear optical devices.

References
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