

Supporting Information for

Universal Switching of Plasmonic Signals using Optical Resonator Modes

Cillian P. T. McPolin^{1,*}, Nicolas Olivier^{1,2,*}, Jean-Sebastien Bouillard^{1,3,*}, Daniel O'Connor^{1,4}, Alexey V. Krasavin¹, Wayne Dickson¹, Gregory A. Wurtz^{1,5,*}, and Anatoly V. Zayats^{1,*}

¹Department of Physics, King's College London, Strand, London WC2R 2LS, UK

²Present address: University of Sheffield, Hounsfield Road, Sheffield S3 7RH, UK

³Present address: University of Hull, Cottingham Road, Hull HU6 7RX, UK

⁴Present address: National Physical Laboratory, Hampton Road, Teddington, Middlesex TW11 0LW, UK

⁵Present address: University of North Florida, 1 UNF Drive, Jacksonville, Florida 32224, USA

*These authors contributed equally to this work.

Correspondence: cillian.mcpolin@kcl.ac.uk

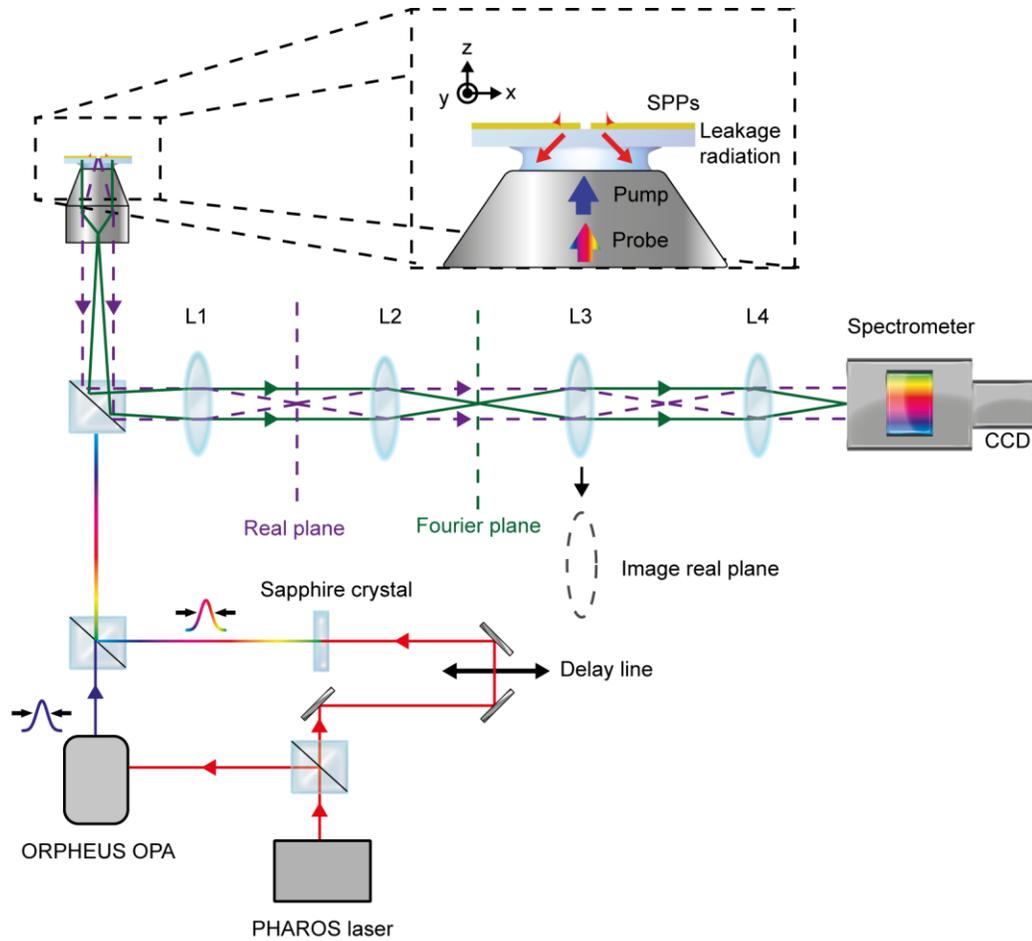


Figure S1. Experimental setup for the pump-probe measurements with Fourier imaging of the leakage radiation.

Movie S1. The experimentally measured variation of the SPP dispersion (a cross-section is shown in Figure 2b) as the upper mirror is moved up and down. The maximum mirror displacement is 220 nm. The cavity modes shift as the mirror is displaced, permitting micromechanical control over the SPP coupling. The structure parameters and experimental conditions are as in Figure 1e.

Supplementary Note 1.

Modeling the Pump-Probe Experiments.

Modeling the nonlinear Kerr-response of gold under pulsed optical excitation was done within the random phase approximation (RPA). In this context, the intensity-dependent permittivity of gold was expressed as the sum of both interband and intraband contributions:

$$\epsilon(\omega) = \epsilon_{\text{inter}}(\omega) + \epsilon_{\text{intra}}(\omega) \quad (\text{S1})$$

where the ϵ_{intra} term relates to free electron motion within a parabolic sp band and ϵ_{inter} relates to interband transitions between a dispersionless d band and the parabolic sp. The intraband contribution was expressed via a Drude-like response:

$$\epsilon_{\text{intra}} = \epsilon_{\infty} - \frac{\omega_p^2}{\omega(\omega + i g_{\text{intra}}(\omega))} \quad (\text{S2})$$

where the plasma frequency is $\omega_p = 2.168 \times 10^{15} \text{ rad s}^{-1}$, ϵ_{∞} is the high frequency permittivity, and $g_{\text{intra}}(\omega)$ characterizes the damping rate of free-electrons. Moreover, the interband term was described within the RPA as:

$$\epsilon_{\text{inter}} = K \int_0^{\infty} \frac{\sqrt{\hbar x - E_g}}{x} [1 - f(x, T_e)] \frac{(\gamma_{\text{inter}}^2 - \omega^2 + x^2) + 2i\omega\gamma_{\text{inter}}}{(\gamma_{\text{inter}}^2 - \omega^2 + x^2)^2 + 4\omega^2\gamma_{\text{inter}}^2} dx \quad (\text{S3})$$

where $K = 1.2695 \times 10^{32}$ is a wavevector independent constant related to the magnitude of the dipole transition moment for the interband transitions. $E_g = 1.98 \text{ eV}$ is the transition energy between the d and sp bands and g_{inter} is the scattering term for interband transitions. The term $f(x, T_e)$ is the Fermi-Dirac distribution for the electrons at an equilibrium temperature T_e .

Once a gold film is optically excited, the electrons respond almost instantaneously, and

the energy subsequently decays via collisions amongst the electrons and lattice phonons, which possess equilibrium temperatures T_e and T_L , respectively. The time response of the electron-phonon interaction was accounted for by the two-temperature model, which relates T_e and T_L via:

$$C_e T_e \frac{\partial T_e}{\partial t} = \nabla \cdot (K_e \nabla T_e) - g(T_e - T_L) + w_0 \text{Im}(\epsilon) \langle \mathbf{E}(r, t) \cdot \mathbf{E}(r, t) \rangle$$

$$C_L \frac{\partial T_L}{\partial t} = g(T_e - T_L) \quad (\text{S4})$$

where $C_e = 67.96 \text{ Jm}^3\text{K}^{-2}$ is the heat capacity of electrons and $g = 2 \times 10^{16} \text{ Wm}^3\text{K}^{-1}$, which is a constant related to the coupling between electrons and phonons. Additionally, $\langle \mathbf{E}(r, t) \cdot \mathbf{E}(r, t) \rangle$ is the time averaged electric field; $K_e = K_{e0} T_e$ is the electron heat diffusion constant with $K_{e0} = 318 \text{ Wm}^{-1}\text{K}^{-1}$ and C_L is the lattice heat capacity.

Thus the two temperature model was solved, together with Maxwell's equations, using COMSOL in order to simulate the pump probe experiment, with the results shown in Figures 4e-g.

Supplementary Note 2.

SPP coupling efficiency definition.

The numerically calculated SPP coupling efficiency (CE) is defined as:

$$CE = P_{SPP} / P_0 \quad (\text{S5})$$

where P_0 is the optical power incident on the slit aperture area and P_{SPP} is the SPP power at the edge of the slit aperture. In order to accurately evaluate P_{SPP} , the SPP power P_x is evaluated at a distance of $x = 10 \mu\text{m}$ from the edge of the slit, taking into account the propagation losses of the

SPP mode:

$$P_{SPP} = P_x \exp(2 \operatorname{Im}(k_{SPP})x) \quad (\text{S6})$$

where $k_{SPP} = k_0 \sqrt{e_d e_m / (e_d + e_m)}$ with e_d and e_m being the frequency-dependent dielectric and metal permittivities, respectively. Using these considerations, we were able to accurately determine a value for the coupling efficiency in the simulations.