Supplementary Material: Giant photo-induced reflectivity modulation of nonlocal resonances in silicon metasurfaces

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Contents

S1 Time-resolved Spectroscopy Setup

In this section we detail the pump-probe setup (see Fig. S1) employed for time-resolved measurements.

The experimental $\Delta R/R$ spectra shown in Fig. 2a of the paper, Fig. 3a of the paper, and Fig. S4 were measured by using a Spectrometer (see Methods). On the other hand, the $\Delta R/R$ dynamics in Fig. S5 were measured by using a Lock-in Amplifier. In such configuration, the reflected probe signal was measured by a single-pixel photodiode, whose output was demodulated by a lock-in amplifier (Ametek DSP 7230) to provide the reflectivity variation signal (δR). The pump beam was modulated by a mechanical chopper wheel, whose operating frequency was employed as the reference for the lock-in amplifier. In parallel with this, the output signal of the photodiode was also measured (without being demodulated) to obtain the signal providing the reflectance at equilibrium (R_{eq}). Finally, a desktop computer acquired both δR and R_{eq} signals and calculated $\Delta R/R = \delta R/R_{eq}$. In this configuration, spectral resolution was accomplished by selecting a portion of the reflected probe spectrum by a band-pass filter (Thorlabs FB-series) mounted in front of the photodiode.

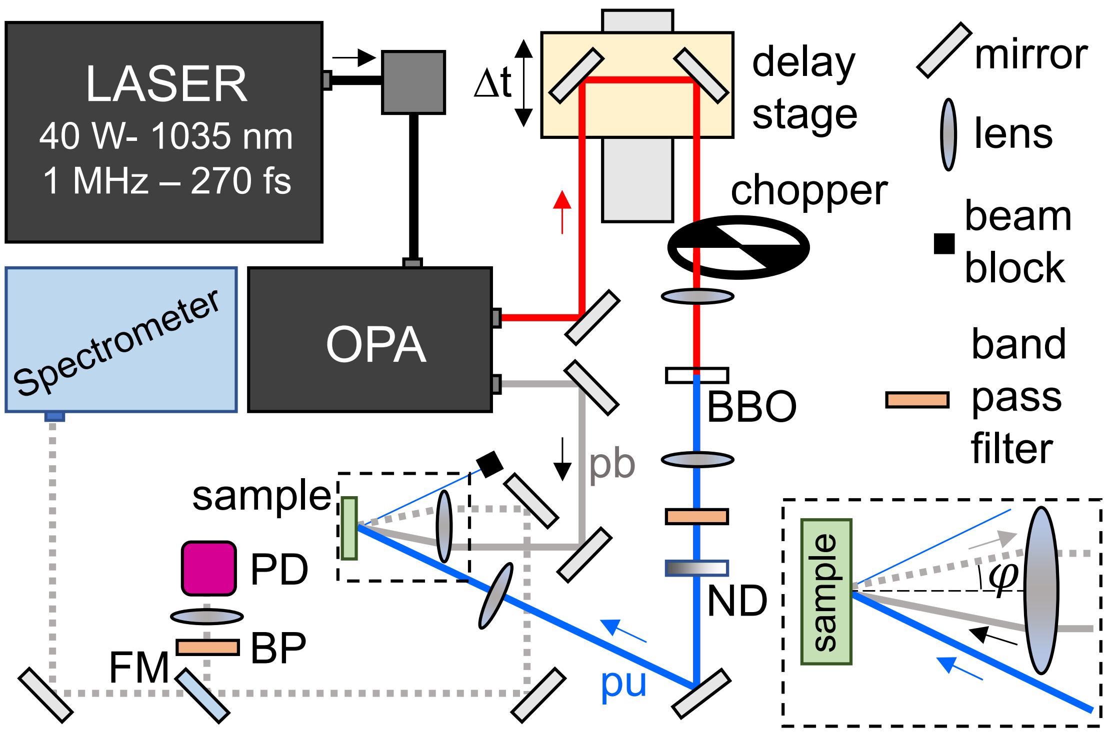

Fig S1 Pump-probe Setup. Schematic of the setup. OPA: optical parametric amplifier; BBO: Barium-Borate crystal. ND: Neutral Density Filter. PD: Photodetector (single-pixel). FM: Flip Mirror. Grey solid (dotted) line: probe beam impinging onto (reflected by) the sample. Red line: signal (SIG) output from the OPA. Blue line: pump beam (second-harmonic radiation from SIG). The inset shows the detailed view of the beams configuration at the sample position (φ is the probe beam angle of incidence).

S2 Time-Resolved Out-of-equilibrium Spectra

In this section we provide an alternative view of the experimental data shown in Fig. 2a. In particular, in Fig. S2, we display the out-of-equilibrium reflectance spectra (R_{out}) at various delay times and we compare them to the at-equilibrium reflectance spectrum (R_{eq}).

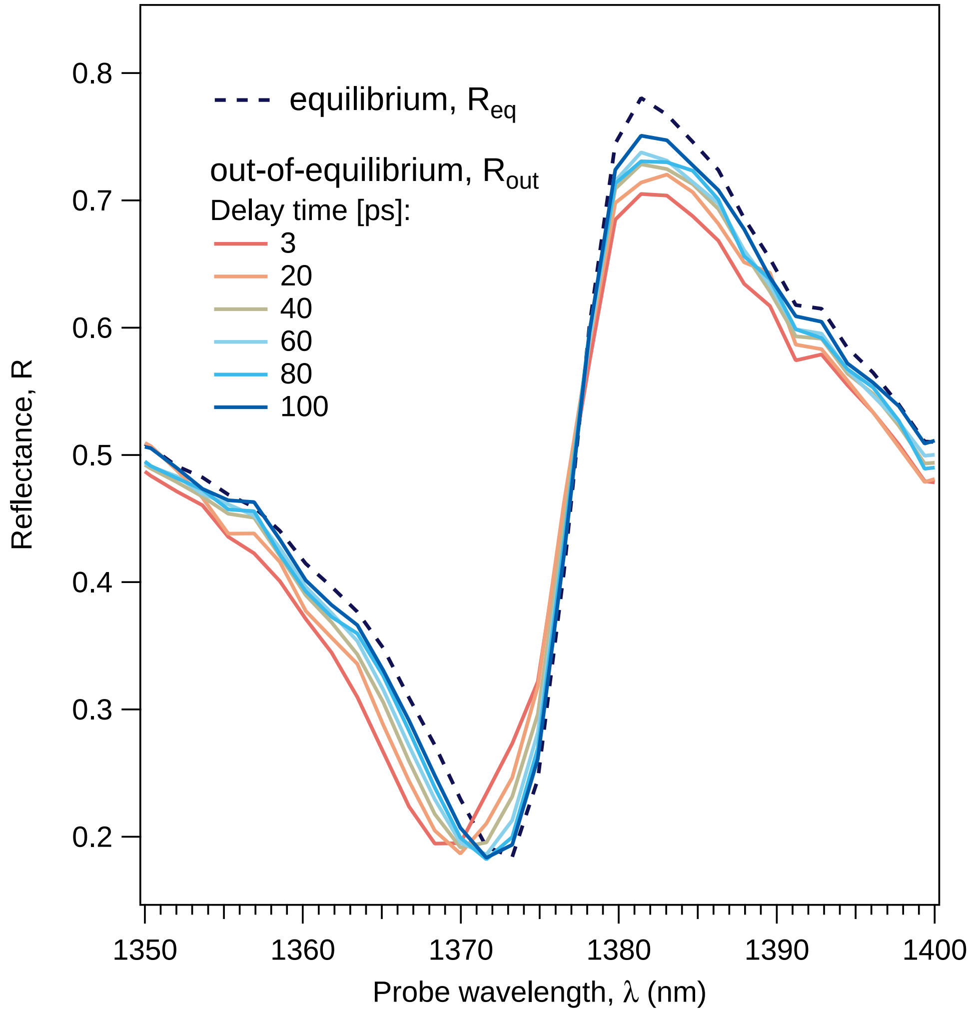

Fig S2 Out-of-equilibrium Reflectance Spectra. Measured reflectance spectra as a function of the probe wavelength: at-equilibrium reflectance R_{eq} (dashed blue line, as in Fig. 1b) and out-of-equilibrium reflectance R_{out} (solid lines, corresponding to the data in Fig. 2a).

S3 Fano-resonance-based Model

In this section, we detail the differential fit procedure we adopted to retrieve the temporal evolution of the out-of-equilibrium spectral position of the GMR by using a Fano lineshape.

In order to properly analyze the equilibrium reflectance data (see Fig. 1b of the paper) by adopting a Fano resonance lineshape,[52] we first converted the linear data from the wavelength λ to the energy domain $E = 2\pi\hbar c/\lambda$ (see blue markers in Fig. S3).

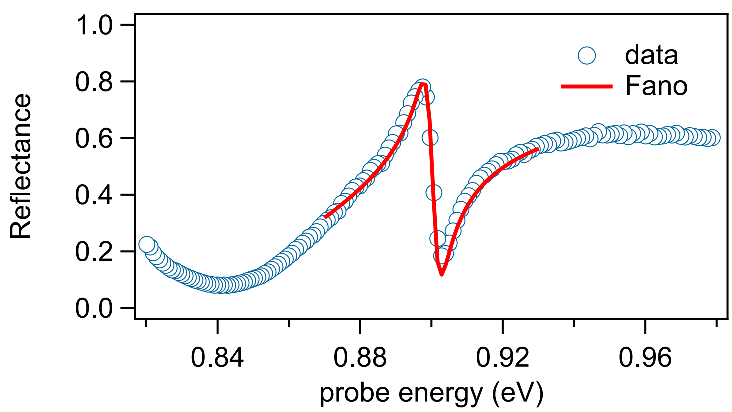

Fig S3 Fano resonance Energy Domain (Equilibrium) Measured reflectance spectra as a function of the probe energy (blue dots) and fit based on Eq. S1 (red line).

Then, to retrieve the equilibrium value of the spectral position ($E_r = 2\pi\hbar c/\lambda_r$), spectral width ($\gamma_r$), quality factor ($Q_F = E_r/\gamma_r$), and profile index ($q_r$) of the Fano resonance, we assumed the equilibrium reflectance to be of the form

$$R_{eq} = R_F\left(E; E_r, \gamma_r, q_r\right) = R_{bkg}(E; E_r) + R_0 + f_0^2 \cdot \frac{\left(q_r \cdot \frac{\gamma_r}{2} + E - E_r\right)^2}{\left(\frac{\gamma_r}{2}\right)^2 + \left(E - E_r\right)^2} \tag{S1}$$

and we fitted Eq. (S1) to the experimental data. In particular, in Eq. (S1), the terms $R_{bkg}(E; E_r)$, R_0, and f_0^2 denote the reflectance background due to the Fabry-Pérot resonance,[29] a constant vertical offset, and amplitude (positive) of the Fano lineshape, respectively. The outcome of the analysis, depicted in Fig. S3, provides $Q_F \sim 170$ and $q_r = -(0.94 \pm 0.02)$.

The measured $\Delta R/R$ spectra (Fig. 2a of the paper), which are displayed also in Fig. S4 in energy domain (markers), have been analyzed by using the differential fit approach. As shown by the theoretical profiles (light blue solid line) in Fig. S4, the experimental data can be reproduced by assuming the out-of-equilibrium signal to be of the form (see Appendix D)

$$\frac{\Delta R}{R} = \frac{R_{out} - R_{eq}}{R_{eq}} \tag{S2}$$

with

$$R_{out} = R_F\left(E; E_r + \Delta E_r, \gamma_r, q_r\right) + \delta R_0, \tag{S3}$$

where ΔE_r and δR_0 represent a variation of the spectral position of the Fano resonance and a variation of the vertical offset, respectively. Therefore, in our approach, the out-of-equilibrium experimental data can be well reproduced by assuming only *two* fitting parameters. Interestingly, our analysis reveals a blue-shift of the resonance spectral position (positive variation of the resonance spectral position in energy domain, $\Delta E_r > 0$). On the other hand, the observed variation of the offset term δR_0 combines with the variation of the Fabri-Pérot background.

Finally, the variation of the resonance spectral position $\Delta\lambda_r$ (in wavelength domain) has been calculated as

$$\Delta\lambda_r = (\lambda_r + \Delta\lambda_r) - \lambda_r = 2\pi\hbar c\left(\frac{1}{E_r + \Delta E_r} - \frac{1}{E_r}\right) = -\frac{2\pi\hbar c}{E_r\left(E_r + \Delta E_r\right)} \cdot \Delta E_r,$$

thus a negative $\Delta\lambda_r$ value.

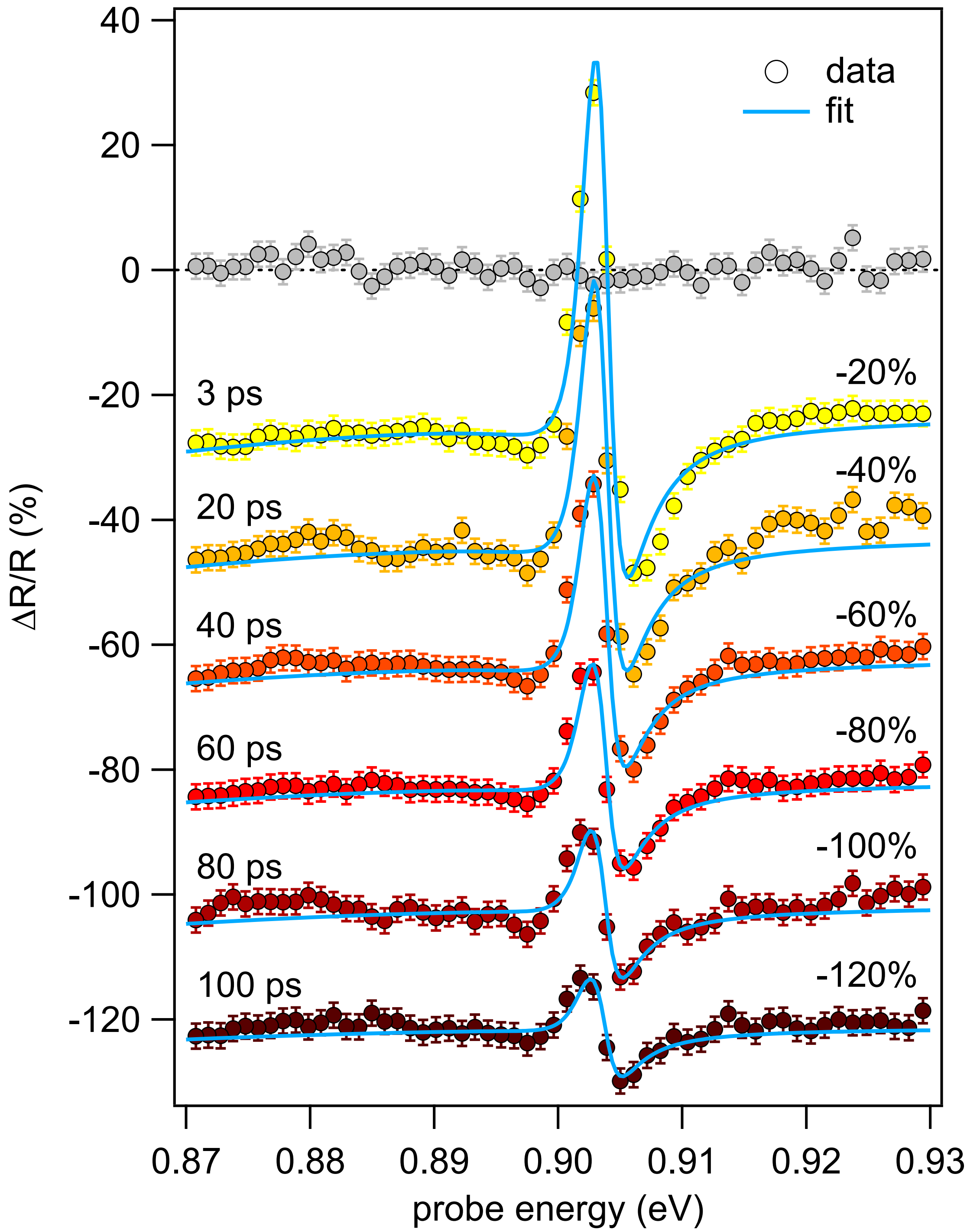

Fig S4 Fano resonance Energy Domain (Out-of-Equilibrium). Analysis (light blue solid line) of the measured (markers) $\Delta R/R$ spectra with a model based on Eq. (S2). The grey dots correspond to the negative time delays, *i.e.*, before the pump pulse arrival. Each spectrum is vertically shifted by the amount indicated by the corresponding label on the right. Labels on the left indicate the Δt value at which the corresponding out-of-equilibrium spectrum has been measured.

S4 Reflectance Modulation for Multilayer Structure

In this section we provide the results of time-resolved pump-probe experiment on a sample consisting in a uniform Si(500 nm)/ SiO_2(1160 nm) bilayer deposited on Si.[29] As a reference, Fig. S5 shows the relative reflectance variation dynamics $\Delta R/R\,(\Delta t)$ measured at 1520 nm probe wavelength. The equilibrium reflectance, measured by the linear spectroscopy setup described in the Methods, is $R_{eq} = 0.2$ at 1520 nm probe wavelength.

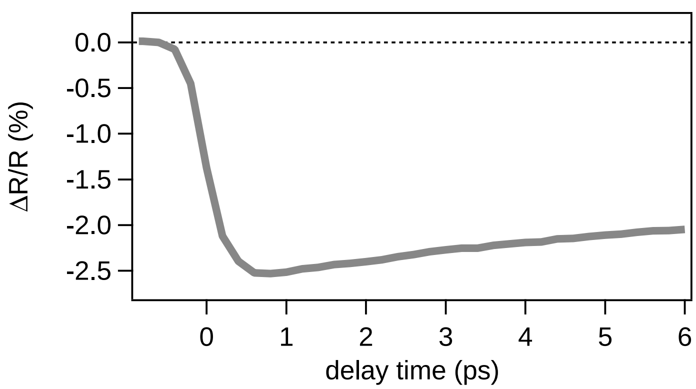

Fig S5 Out-of-equlibrium Dynamics of Multi-layer. Relative reflectance variation as a function of delay time at 1520 nm probe wavelength obtained for a Si(500 nm)/SiO_2(1160 nm)/Si bilayer structure at pump fluence $F_{inc} = (120 \pm 20)$ μJ/cm^2.