Supplementary Information:

Supplementary Figures

**Supplementary Figure 1 | Simulation of 120x120 Swiss cross without material loss.** To understand the role of radiation damping we simulate the Swiss cross nanostructure on a substrate in a periodic lattice with no material damping. We observe quality factors of 740 and 7500 for the lower and higher frequency symmetric modes. These quality factors are significantly higher than for a sphere embedded in a medium (Q≈10), indicating much lower radiation damping due to weaker mechanical coupling.

**Supplementary Figure 2 | Pump and probe wavelength relative to the resonances of the 120x90 nm nanostructure array.** The pump is on the long wavelength side of both vertical and horizontal plasmon resonances. The probe pulse is on the long wavelength side of the vertical resonance and the short wavelength side of the horizontal resonance. The sign change of the electronic coincidence when the probe polarization is rotated (insets of Fig 1 d and f) is one prominent consequence of the probe switching between the red and blue sides of the plasmon resonance.
**Supplementary Figure 3 | Simulation procedure.** The phonon dynamics are simulated in the frequency domain using the finite element solver COMSOL Multiphysics. (a) First, the phonon excitation is simulated by calculating the thermal expansion of the nanoparticle due to a raised temperature, relative to that of the substrate. The temperature increase causes a strain proportional to the temperature change and the coefficient of thermal expansion of the gold $\alpha = \frac{\Delta T}{T}$. The distorted geometry is shown in (a) with a color scale to indicate the magnitude of the displacement from the original geometry, where red indicates a larger displacement. (b) The displacement is then calculated for each frequency given the thermal stress. We use periodic boundary conditions on the outer boundaries and a perfectly matched layer on the bottom to absorb propagating acoustic waves. (c) The detection of the phonon by the plasmon is simulated by assuming the shift in the plasmon resonance, and thus the amplitude of the differential transmission, is proportional to the change in length along the axis parallel to the probe polarization. The length change is calculated by integrating the displacement parallel to the probe polarization across that face of the nanostructure.

**Supplementary Figure 4 | Nanomechanical simulations of the phononic response of isolated metallic nanostructures (blue) and nanostructures in contact with a fused silica substrate (green).** An isolated bar (a,b, blue) has one resonance (extensional mode) and two when coupled to a substrate (a,b, green). An isolated Swiss-cross (c,d, blue) with non-equal arms has two modes, the symmetric and anti-symmetric modes. When coupled to the substrate (c,d, green), more modes are excited. In both cases, changing the polarization of the detection plasmon from vertical to horizontal produces the characteristic interference as seen in the experiments (Fig 1). In the isolated symmetric cross (e,f, blue), only the symmetric mode is excited due to the symmetry of the excitation process, despite the anti-symmetric mode being an eigenmode of the system. A loss factor of $\eta = 0.1$ was used to account for the energy loss in the gold and inhomogeneous broadening. The largest amplitude peaks are not affected by the lattice period and thus represent localized acoustic vibrations rather than surface acoustic waves.
Supplementary Figure 5 | Substrate effect on the 120x120 nm Swiss cross. To demonstrate the effect of the substrate on the phonon modes, we simulate the mechanical motion as a function of frequency for different coupling strengths between the nanostructure and the substrate. The coupling is controlled by a thin elastic layer with a spring constant per unit area varying from 0.1 GPa/nm to 1000 GPa/nm. In the weak coupling limit the dynamics are the same as without a substrate, while in the limit of strongest coupling the dynamics are the same as for a rigid connection. (a) The absolute value of the displacement of the x (or y from symmetry) axis of the nanostructure is plotted as a function of substrate coupling and frequency. (b) A cross section for the weakest (blue) and strongest (purple) coupling shows a number of peaks which are mechanical eigenmodes. For weak coupling (blue), only one eigenmode exists (in this frequency range) which corresponds to an in phase stretching of the “arms”. The mechanical displacement for this mode is shown in (c), where the black lines represent the original geometry and the solid surface is the distorted geometry, with the color indicating the magnitude of the displacement. When the coupling strength increases, the in plane mode couples with the out of plane modes, creating two new modes, where the in and out of plane motion has different phase relationships as seen in (d) and (e), which is supported by the anti-crossing seen in (a).
Supplementary Figure 6 | Substrate effect on the 120x90 nm Swiss cross. To demonstrate the effect of the substrate on the phonon modes, we simulate the mechanical motion as a function of frequency for different coupling strengths between the nanostructure and the substrate. The coupling is controlled by a thin elastic layer with a spring constant per unit area varying from 0.1 GPa/nm to 1000 GPa/nm. In the weak coupling limit the dynamics are the same as without a substrate, while in the limit of strongest coupling the dynamics are the same as for a rigid connection. (a) The absolute value of the displacement of the x and y axes of the nanostructure is plotted as a function of substrate coupling and frequency. (b) A cross section for the weakest (blue) and strongest (purple) coupling shows a number of peaks which are mechanical eigenmodes. For weak coupling (blue), only two eigenmodes exists (in this frequency range) which corresponds to an out of phase stretching of the “arms” (e) and an in phase stretching of the “arms” (f). The mechanical displacement for the modes are shown in (e–j), where the black lines represent the original geometry and the solid surface is the distorted geometry, with the color indicating the magnitude of the displacement. When the coupling strength increases, the in plane modes couples with the out of plane modes, creating new modes, where the in and out of plane motion has different phase relationships as seen in (g) and (h) for the anti-symmetric mode and (i) and (j) for the symmetric mode.
**Supplementary Figure 7 | Schematic of experiment and example result** (a) We measure the phonon dynamics in plasmonic nanostructures with a pump-probe experiment. A pump and control pulse with a varying time delay excite plasmons in the Swiss-cross nanostructures. The plasmons generate phonons which are observed using a probe pulse: producing a time dependent transmission (b). The Fourier transform (c) of the transient transmission has several peaks associated with the phonon modes of the nanostructures, which allow us to probe displacements at the nanoscale at GHz frequencies.

**Supplementary Figure 8 | Data analysis procedure.** The differential transmission \((\Delta T)\) is the pump induced change in the sample transmission and is acquired by measuring the transmission of a probe pulse through the sample while modulating the pump pulse. The differential transmission as a function of time delay between the pump and probe pulses is plotted for a horizontally polarized probe pulse in (a), and a vertically polarized probe pulse in (b). The time trace is then differentiated (c and d) to remove the slowly varying background and Fourier transformed to obtain the differential transmission as a function of frequency. The temporal window for the Fourier transform is chosen to isolate the contribution from the phonons and stretches from the end of the electronic coincidence to after the phonons have decayed (10 ps – 1500 ps). The Fourier transform is calculated with the 10 ps – 1500 ps window (red) and with a shorter window (blue-dashed) to demonstrate that the features observed are robust to window choice. The interference feature in the Fourier transform of the horizontally polarized data (e) is present with both window choices.
**Supplementary Figure 9** | **Analytic and numerical solutions for a gold sphere in a glass matrix.** We calculate the dynamics of a 40 nm gold sphere in response to an impulsive thermal strain. (a) A schematic of the system consisting of a sphere in the center of an infinite glass medium. (b) The radial displacement at the surface shows a resonance corresponding to the phonon mode. The analytic and numerical solutions both give a resonant frequency of 39.59 GHz and a quality factor of 9.4. These values are also in reasonable agreement with the approximate resonance frequency and quality factor given by the simple relations\(^1\): \( f \approx \frac{v_p}{(2R)} \approx 40.5 \text{ GHz} \) and \( Q \approx \frac{\pi \rho_{Au}}{\rho_{Si} v_p} / \left( \frac{2 \rho_{Si} v_p}{\rho_{Si} v_p} \right) \approx 7.49 \). (c) The real (blue curve) and imaginary (purple curve) displacement as a function of distance from the sphere, plotted at the resonance frequency, shows strong oscillations in the glass matrix (blue region) as well as in the nanoparticle (gold region).

**Supplementary Figure 10** | **Magnitude, phase, and amplitude of the transient transmission for the 120x90 nm Swiss cross.** The magnitude of the Fourier transform for the horizontal (a) and vertical (b) probe. The phase of the Fourier transform for a horizontal (c) and vertical probe (d). The amplitude of the real (blue) and imaginary (purple) components of the Fourier transform for a horizontal (e) and vertical (f) probe. For the vertical probe, the overall sign of the FT was inverted to account for probing on the opposite side of the plasmon resonance from the horizontally polarized probe.
Supplementary Figure 11 | Polarization dependence of the transient reflectivity for the 120x120 nm Swiss cross. Similar amplitudes and time dependences are observed. The small differences in the amplitude between pump and probe parallel (pumpH-probeH) and perpendicular (pumpV-probeH) are attributed to fabrication asymmetries, resulting in slightly different plasmon resonances and thus coupling efficiencies.
Supplementary Tables

**Supplementary Table 1**: Parameters used in numerical simulations from Ref\textsuperscript{2}.

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Supplementary Notes

Supplementary Note 1

We now detail the function used in Figure 3 to fit the frequency domain response of the phonons. The phonon creates a periodic modulation of the transmission which decays with time. We assume the modulation of the transmission is of the form:

\[ n(t, \tau) = ae^{-\gamma(t-\tau)} \cos(\omega_0(t-\tau))\theta(t-\tau) \]

where \( \theta(t-\tau) \) is the Heaviside function which ensures the response occurs only after the pump strikes the sample at time \( \tau \), \( \gamma \) is the decay rate, \( \omega_0 \) is the frequency, and \( a \) is the amplitude of the phonon mode. The inverse fourier transform of \( n(t, \tau) \) is then:

\[ \hat{n}(\omega, \tau) = \frac{ie^{-i\omega \tau}}{2\pi} \frac{(\omega-i\gamma_0)}{\omega_0^2-(\omega-i\gamma_0)^2} \]

we are considering a pump probe experiment with two pumps, so we are summing two functions, one for the first pump and one for the second with a time delay of \( \tau \) and considering the signal after the second pump pulse:

\[ n_{\text{pump-pump}}(t, \tau) = ae^{-\gamma t} \cos(\omega_0(t))\theta(t-\tau) + ae^{-\gamma(t-\tau)} \cos(\omega_0(t-\tau))\theta(t-\tau) \]

The IFT is then

\[ \hat{n}_{\text{pump-pump}}(\omega, \tau) = \frac{ie^{-i\omega \tau}}{2\pi} \frac{\left[(i\omega+\gamma_0)\cos(\omega_0 t)-\omega_0 \sin(\omega_0 t)\right]e^{-\gamma_0 \omega} + (i\omega+\gamma_0)}{\omega_0^2-(\omega-i\gamma)^2} \]

For data analysis purposes, the derivative of the transient transmission is used, which can be obtained by multiplying the IFT by \( i\omega \):

\[ \hat{n}'_{\text{pump-pump}}(\omega, \tau) = \frac{ai\omega e^{-i\omega \tau}}{2\pi} \frac{\left[(i\omega+\gamma_0)\cos(\omega_0 t)-\omega_0 \sin(\omega_0 t)\right]e^{-\gamma_0 \omega} + (i\omega+\gamma_0)}{\omega_0^2-(\omega-i\gamma)^2} \]

This function is then used to model the phononic response under multiple-pump excitation as shown in Figure 3.
Supplementary References