

Supporting information for the paper

Controlling the Quality Factor of a Single Acoustic Nanoresonator by Tuning its Morphology

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File content:

- Description of finite-element simulations.
- Figure S1: Oscillating components of time-resolved signals measured for a variety of ND aspect ratios (examples).
- Figure S2: Geometry considered in the FEM simulations.
- Figure S3: FEM-computed excitation vibrational spectra for different ND aspect ratios.
- Figure S4: Excitation vibrational spectrum computed for $\eta=2$ ND aspect ratio (zoom).

Description of finite-element modeling (FEM) simulations

FEM simulations based on the numerical resolution of Navier equation of elasticity were performed using the Structural Mechanics module of the COMSOL commercial software, adapting the general strategy to include nano-object environment described in a previous work¹ to the experimental situation investigated here. A simulation domain consisting of a cylindrical ND and its underlying plane substrate was considered (Fig. S2). Materials were described by their elastic constants (Young modulus Y and Poisson ratio ν) and density d . $Y_{Au}=78$ GPa, $\nu_{Au}=0.44$ and $d_{Au}=19300$ kg.m⁻³ values were used for gold, and $Y_{Sa}=300$ GPa, $\nu_{Sa}=0.22$ and $d_{Sa}=3900$ kg.m⁻³ for sapphire. A perfect mechanical contact (i.e., continuity of displacement and normal stress) was assumed at the ND/substrate interface, and stress-free boundary conditions were used on the other ND faces. No intrinsic acoustic damping mechanism was considered in the ND and its environment (i.e., infinite Q_{int} , so that the simulated quality factors correspond to Q_{env}). Spherical perfectly matched layers (PMLs) were used at the border of the simulation domain to avoid spurious reflections of acoustic waves.

Simulations were performed in the frequency domain, considering a periodical mechanical excitation of the ND (a uniform stress corresponding to the experimental case of a homogeneously excited ND being chosen; note that this choice affects the mode amplitudes, but not their frequencies and quality factors). Simulated excitation vibrational spectra such as those shown in Fig. 5 of the main text were deduced from the computed displacement profiles by plotting the average elastic energy stored in a ND as a function of excitation frequency. Since both the cylindrical ND geometry and the exciting stress display radial symmetry about the vertical axis containing the ND center, FEM simulations could be performed in a 2D axisymmetric geometry instead of a 3D one, greatly reducing the time and memory required for the calculations. Excited vibrational modes appear as Lorentzian resonances in the computed spectra (Fig. 5 of the main text), allowing the determination of their associated frequency, spectral width and quality factor, as well as of the total elastic energy they store (the resonance spectral area allowing to compare the efficiency with which different vibrational modes are expected to be excited in the context of time-resolved experiments).

Numerical simulation parameters (mesh size, PML position and thickness) were optimized through a series of preliminary computations on nanospheres embedded in a homogeneous elastic (or viscoelastic) environment, a system for which analytical results are available.^{2,3} In particular, the choice of a mesh size much smaller than ND size appeared critical for an accurate prediction of vibrational quality factors. A maximum element size of one tenth of ND height was typically used in this work.

References

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- (3) Galstyan, V.; Pak, O. S.; Stone, H. A. A Note on the Breathing Mode of an Elastic Sphere in Newtonian and Complex Fluids. *Phys. Fluids* **2015**, *27*, 32001.

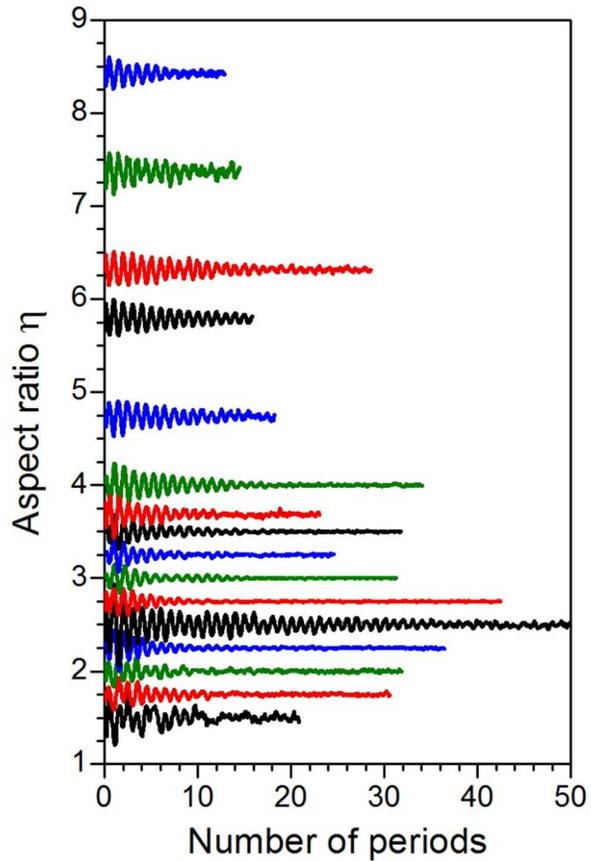


Figure S1. Oscillating components of time-resolved pump-probe transmission changes $\Delta T/T$, measured for a variety of ND aspect ratios (left axis), using in each case a probe wavelength in the ND localized surface plasmon resonance range (Fig. 1) and a pump wavelength away from it. Signals are normalized in amplitude and vertically shifted for comparison. The time unit was defined as the period of the detected mode with the highest quality factor.

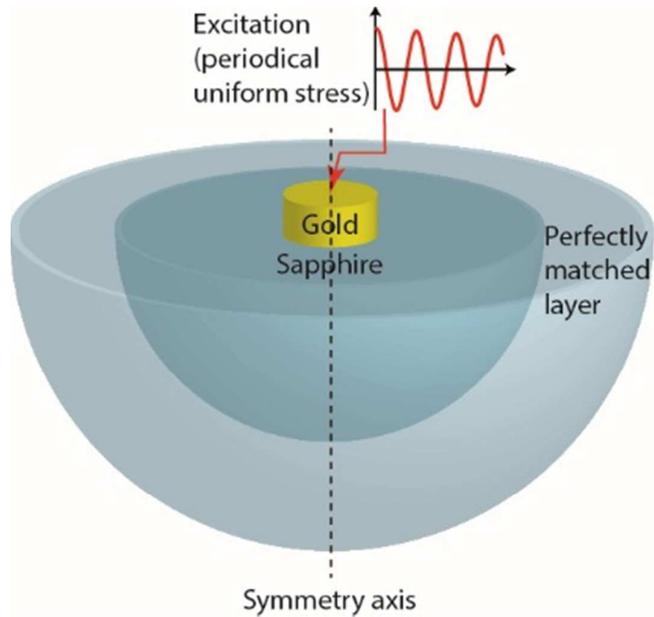


Figure S2. Geometry considered in the FEM simulations: cylindrical gold ND on top of a sapphire substrate, modeled as a hemisphere surrounded by a perfectly matched layer. A periodical mechanical excitation of the ND is assumed. The presence of a symmetry axis enables to perform two-dimensional calculations instead of three-dimensional ones.

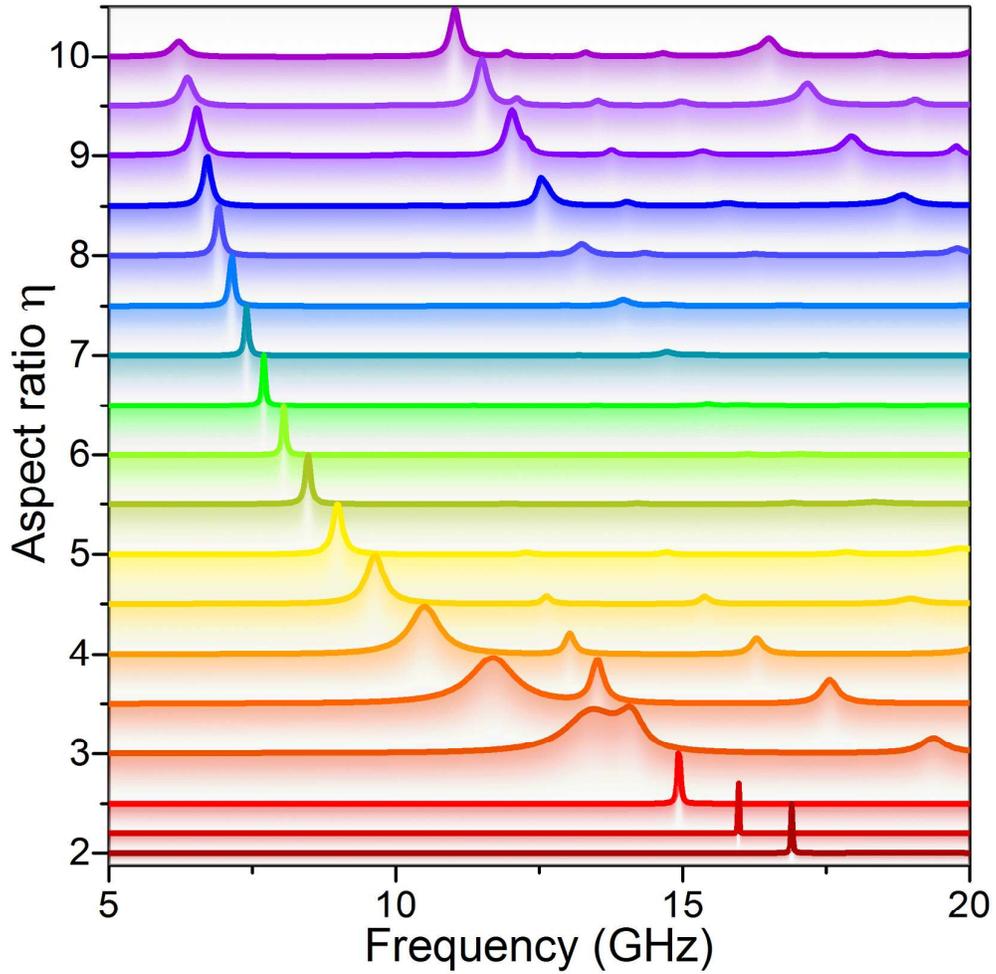


Figure S3. FEM-computed excitation vibrational spectra for different ND aspect ratios. The average elastic energy stored in the ND (normalized) as a result of a periodic ND excitation (with uniform isotropic stress) is plotted as a function of the excitation frequency. Note that the low normalized amplitude of some weakly excited and/or strongly damped modes makes them undistinguishable on this figure, a zoom being required for their observation (as shown for the $\eta=2$ case in Fig. S4).

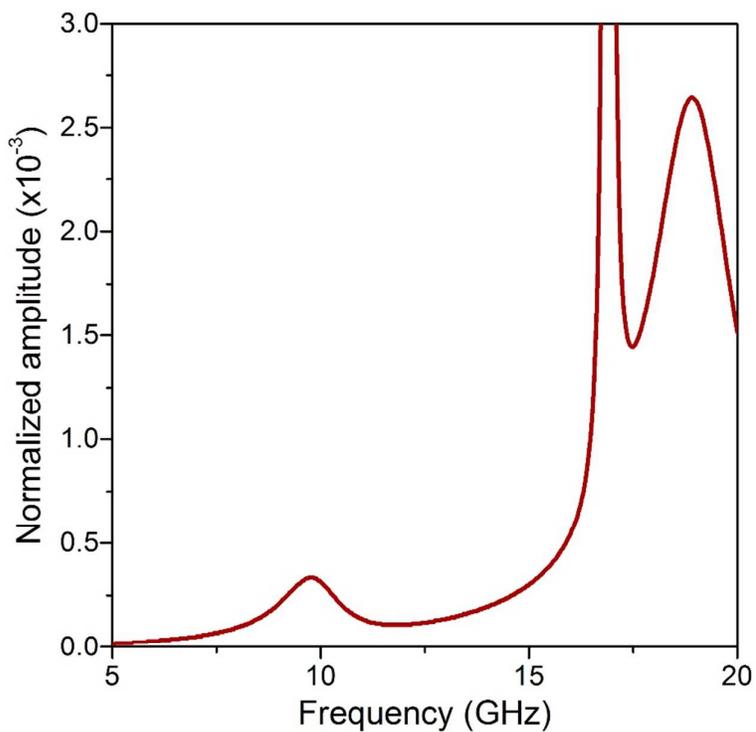


Figure S4. Excitation vibrational spectrum computed for $\eta=2$ ND aspect ratio (zoom). This zoom makes visible modes at 9.8 and 18.9 GHz frequencies having much larger spectral widths and much lower normalized amplitudes than those of the 16.9 GHz mode already easily distinguishable in Fig. S3.