

Ultrastrong plasmon-phonon coupling via epsilon-near-zero nanocavities

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ABSTRACT

Vibrational ultrastrong coupling (USC), where the light-matter coupling strength is comparable to the vibrational frequency of molecules, presents new opportunities to probe the interactions of molecules with zero-point fluctuations, harness cavity-modified chemical reactions, and develop novel devices in the mid-infrared spectral range. Here we use epsilon-near-zero nanocavities filled with a model polar medium (SiO₂) to demonstrate USC between phonons and gap plasmons. We present classical and quantum mechanical models to quantitatively describe the observed plasmon-phonon USC phenomena and demonstrate a modal splitting of up to 50% of the resonant frequency (normalized coupling strength $\eta > 0.25$). Our wafer-scale nanocavity platform will enable a broad range of vibrational transitions to be harnessed for USC applications.

Main

The field of cavity quantum electrodynamics (QED) has generally divided light-matter interactions into two regimes: weak coupling (WC), when losses exceed the light-matter coupling strength, and strong coupling (SC), when the coupling strength dominates^{1,2}. For weak-coupling phenomena such as the Purcell effect³, Fano interference⁴, and surface-enhanced infrared absorption⁵, the coupled systems exchange energy on a time scale slower than the decay rates. In contrast, within the SC regime, the oscillators exchange their energy reversibly and coherently over an extended time frame that is longer than the decay rates⁶⁻¹⁴, enabling applications such as quantum information processing¹⁵. Furthermore, strong coupling leads to the formation of hybridized energy states that are different from those of the bare constituents, leading to novel phenomena such as the modification of chemical reaction rates^{2,16,17} and of ground-state reactivity¹⁸.

Even more exotic phenomena can occur when the normalized coupling strength, η , of the system exceeds ~ 0.1 , where η is defined as the ratio of the light-matter coupling strength, g , to the mid-gap frequency, ω . In this ultrastrong coupling (USC) regime^{1,2,19,20}, some of the standard approximations that are valid for WC and SC, such as the rotating-wave approximation, are expected to break down. Furthermore, transitioning from SC to USC implies that the hybrid mode exhibits substantially more oscillations between light and matter states prior to decay, and such fast and efficient interactions can enable novel ultrafast devices^{19,21}. Another striking phenomenon predicted in the USC regime, resulting from the antiresonant term in the light-matter coupling equation, is the possibility to extract virtual photons from the modified ground state via a dynamic Casimir effect^{22,23}. Finally, USC between light and molecules has the potential to modify or enhance chemical reactions beyond what is possible in the SC regime¹⁹.

USC has been demonstrated using photochromic molecules²⁴, circuit QED systems²⁵, intersubband polaritons²⁶, molecular liquids²⁷, and two-dimensional electron gases²⁸. SC to vibrational modes at mid-infrared (MIR) frequencies has been demonstrated in various systems^{18,29-32}, enabling applications such as surface-enhanced vibrational spectroscopy^{5,33}, thermal emission and signature control, and modified heat transfer. It has been challenging, however, to achieve USC at MIR frequencies, particularly in solid-state systems, because of the relatively weak oscillator strengths for vibrational modes originating from

lattice ionic motions. Previous demonstrations have involved extended microcavity structures^{2,18,27}, opening up a new physical regime at these technologically important frequencies, but with limited possibilities for novel nonlinear effects.

Here we demonstrate vibrational USC within nanocavities, drastically reducing the size of the system and thus the amount of material involved in achieving MIR USC. Specifically, we use coaxial nanocavities, which exhibit strong transmission resonances and field enhancements close to the cutoff frequency of the TE₁₁-like mode; these can be understood as resulting from excitation of the zeroth-order Fabry-Perot resonance³⁴ or, alternatively, as arising from an effective epsilon-near-zero (ENZ) effect^{35–37}. We couple this nano-coax ENZ mode to the lattice vibrations of SiO₂ and demonstrate MIR USC with a mode splitting exceeding 50% of the vibrational transition frequency ($\eta > 0.25$) within a solid-state system.

Results

Design and fabrication of ENZ nanocavities

In our USC platform, SiO₂-filled coaxial nanoapertures fabricated into a metal (Au) film are hexagonally arranged with varied gap size (G), diameter (D), and lattice periodicity (P) (Fig. 1a). Unlike conventional approaches of selectively etching annular gaps in metal films, we adopt a novel fabrication approach called atomic layer lithography^{36,38} to create dielectric-filled nanogaps in metal films (Fig. 1c and Method section). After employing standard photolithography to define gold disk arrays on a silicon wafer, SiO₂ films are conformally grown via atomic layer deposition³⁸ (ALD) on the exposed surfaces and sidewalls, followed by subsequent metal cladding layer deposition and planarization via glancing-angle ion milling. This batch process can produce wafer-sized arrays of coaxial apertures with a gap size down to 1 nm, limited only by the ALD growth.

For very narrow gaps, TE₁₁ coaxial modes have a strong plasmonic character. Such modes are characterized by an effective dielectric constant that is approximately zero at cutoff. The resonance frequency of the cavities can be shifted toward longer wavelengths without sacrificing modal confinement by increasing the coax diameter without changing the gap width. In addition, the very long wavelength associated with near-zero permittivity produces a spatially uniform optical field (Fig. 1b lower panel), which in turn provides efficient coupling to the material within the gaps.

We note that the ENZ resonance is a single-aperture effect; in other words, the existence of an array is not required for the transmission resonance we utilize (Sec. S2). We also note that the resonant optical properties of the nanocavities are robust against process variations and inhomogeneous broadening for two reasons. First, the critical dimension (i.e. the width of the nanogap) is lithography-independent and precisely defined by the thickness of the SiO₂ film grown by ALD. Second, the ENZ mode (i.e., the zeroth-order Fabry-Perot resonance) is independent of the cavity length, and is thus insensitive to variations in the gold film thickness.

We designed a series of cavities with diameters ranging from 430 nm to 1120 nm in 30 nm steps, in order to tune the bare ENZ resonance from approximately 1000 to 2500 cm⁻¹. This range is chosen to cover the Reststrahlen band of SiO₂, which is defined by the transverse optical (TO) and longitudinal optical (LO) phonon frequencies (Fig. 2a). Coaxial nanocavities at the ENZ condition show extraordinary optical transmission (EOT)³⁹, since incident light with wavelengths 4–10 μ m can pass through annular gaps that are over thousand times narrower than the wavelength, with the absolute transmittance as high as 60%.

Transmission spectra through the nanocavity arrays were measured by far-field Fourier transform infrared (FTIR) spectroscopy over a large-area (5 mm $\times$ 5 mm) chip containing arrays of coaxial nanocavities. The normalized transmission spectra for coaxial nanocavities filled with 21 nm-thick SiO₂ are plotted in Fig. 2b. The observed transmission features a clear anticrossing behaviour characteristic of SC.

Although these results suggest that the SiO₂ phonons and cavity photons are strongly coupled, the split transmission peaks could potentially be interpreted as the result of a single broad transmission resonance being quenched in a central frequency range by the strong vibrational absorption close to ω_{TO} . To clarify the origin of the split transmission peaks, we perform a theoretical analysis of the coupled system to be compared to our experimental results.

Theoretical analysis of ultrastrong plasmon-phonon coupling

Classical description

Electromagnetic propagation in waveguides is usually based on a description where the phononic degrees of freedom have been integrated out, providing an effective medium $\epsilon(\omega)$ for the propagation of the photon (Sec. S3). However, in order to determine whether the system is in the SC regime, it is essential to retain both phononic and photonic degrees of freedom. We therefore consider a given waveguide mode M , characterized by a wavevector k along the waveguide axis (in this case, $M=TE_{11}$, the fundamental mode of the coaxial waveguide). When the aperture is filled with a uniform dielectric constant ϵ_∞ (originating from coupling to electronic degrees of freedom), the electric field, $\mathbf{E} = E\mathbf{E}_M$, satisfies the wave equation

$$\nabla \times \nabla \times E\mathbf{E}_M - \epsilon_\infty \frac{\omega_k^2}{c^2} E\mathbf{E}_M = 0, \quad (1)$$

where E is the field amplitude and $\mathbf{E}_M$ is a normalized transverse solution of Maxwell's equations (Sec. S4). When the phononic material fills the waveguide, we assume a local relation (valid for non-dispersive vibrational modes) between $\mathbf{E}$ and the relative displacement of ions, $\mathbf{x}$, following Ref. 40. Then, for a given mode profile, $\mathbf{E}(\mathbf{r}) = E \mathbf{E}_M(\mathbf{r}, k)$ and $\mathbf{x}(\mathbf{r}) = x \mathbf{E}_M(\mathbf{r}, k)$, with

$$\ddot{x} = \gamma_{11}x + \gamma_{12}E, \quad (2)$$

$$P = \gamma_{12}x + \gamma_{22}E, \quad (3)$$

where P is the amplitude of polarization vector $\mathbf{P} = P \mathbf{E}_M$, $\gamma_{12}^2 = \omega_p^2 \epsilon_\infty / 4\pi$ is the coupling constant, $\omega_p^2 = \omega_{LO}^2 - \omega_{TO}^2$, $\gamma_{11} = -\omega_{TO}^2$, and $\gamma_{22} = (\epsilon_\infty - 1)/4\pi$ (see Sec. S3). Note that γ_{22} incorporates the effect of the electronic resonances excited at higher frequencies. The wave equation becomes, in this case,

$$\nabla \times \nabla \times E \mathbf{E}_M - \frac{\omega^2}{c^2} (E + 4\pi P) \mathbf{E}_M = 0. \quad (4)$$

By inspection, we find that $\mathbf{E}_M(\mathbf{r}, k)$ satisfies Eq. (4) if $\omega^2(E + 4\pi P) = \epsilon_\infty \omega_k^2 E$ (strictly speaking, we are neglecting the variation between ω and ω_k in the impedance of the metal surrounding the waveguide; for a deeper analysis of how this affects the cutoff condition, see Secs. S2 and S5). This condition, together with Eqs. (2) and (3), can be expressed in a matrix form, stating that $\mathbf{E}_M(\mathbf{r}, k)$ is still a solution of Maxwell equations but at a frequency ω satisfying

$$\begin{pmatrix} \omega^2 - \omega_{TO}^2 & \omega \omega_p \\ \omega \omega_p & \omega^2 - \omega_k^2 \end{pmatrix} \cdot \begin{pmatrix} \omega x \\ \sqrt{\epsilon_\infty / 4\pi} E \end{pmatrix} = 0. \quad (5)$$

(See Sec. S6 for a detailed derivation and a discussion of the effects of losses) Note that the simpler coupled-harmonic-oscillator model that is commonly used for fitting experimental data (see Sec. S7) does not show the ω -dependence of the off-diagonal terms and therefore cannot describe the polaritonic branches correctly (cf. Figs. 3a and S6).

Quantum-mechanical description

We have used a classical approach above. For future work, especially the investigation of nonlinear optical effects, a full quantum mechanical description will be required, wherein off-resonant terms may be important and the rotating-wave approximation cannot necessarily be applied^{41–45}. We therefore apply the canonical procedure of second quantization to the classical Hamiltonian (see Sec. S9 for details). For the sake of convenience, absorption is neglected and the walls of the coaxial cavity are assumed to be perfectly conducting. We obtain a Hopfield-like Hamiltonian⁴¹ for interacting photons and phonons: $H = H_{photon} + H_{phonon} + H_{int}$, with

$$H_{photon} = \sum_m \hbar \omega_m \left(a_m^\dagger a_m + \frac{1}{2} \right), \quad (6)$$

$$H_{phonon} = \sum_m \hbar \omega_{TO} \left(b_m^\dagger b_m + \frac{1}{2} \right), \quad (7)$$

$$H_{int} = \sum_m \hbar \left[i C_m (a_m^\dagger b_m - a_m b_m^\dagger) + D_m (2 a_m^\dagger a_m + 1) \right. \quad (8)$$

$$\left. + i C_m (a_m b_m - a_m^\dagger b_m^\dagger) + D_m (a_m a_m + a_m^\dagger a_m^\dagger) \right]. \quad (9)$$

where a^+ (a) and b^+ (b) are the creation (annihilation) operators for photons and phonons, respectively, the sum is over modes m , and

$$C_m = \frac{\omega_p}{2} \sqrt{\frac{\omega_{TO}}{\omega_m}}, \quad D_m = \frac{\omega_p^2}{4\omega_m}. \quad (10)$$

As expected, given that the system comprises two coupled harmonic oscillators, diagonalizing the full Hamiltonian gives eigenfrequencies identical to those obtained from Eq. 5. In other words, the classical approach can be used to compute the polaritonic branches exactly when the system enters into the USC regime. The same behaviour is valid for an arbitrary number of vibrational modes in the polar material (see Secs. S8, S10).

Experimental verification of dispersion and ultrastrong coupling

We can therefore analyse the dependence of the ENZ resonant frequencies on the coax inner diameter using Eq. 5 (Fig. 3 c-f). As ω_p is independent of the hole shape and size, the ENZ resonances in this SC condition depend only on the bare ENZ frequency, $\omega_{ENZ}(D)$. This bare ENZ frequency, in turn, is obtained from the upper polariton branch (see Sec. S12). This procedure is followed because the lower polariton is more affected by additional vibrational resonances that have not been considered in this simple analysis.

For each gap size, we estimate the coupling strength g as half the frequency difference between the upper and lower polariton branches when $\omega_{ENZ}(D) = \omega_{TO}$. Results are summarized in Figure 3b and Table 1. Since the SiO₂ layer is added to our coaxial nanocavity during the fabrication process, it is not trivial to experimentally measure the bare ENZ resonance (i.e., to measure the transmission of the cavity with an empty gap). Instead, we measure the linewidth of a bare ENZ resonance at a higher frequency, where it is uncoupled from the SiO₂ phonons. As shown in Table 1, all cavities exhibit modal splittings that are larger than the average of the linewidths of the bare cavity resonances and of the TO phonon ($2g/\gamma_{avg} > 1$). This indicates that the system is in the SC regime.

Moreover, the normalized coupling strength $\eta = g/\omega_{TO} > 0.25$, regardless of gap width, indicating that the systems are also in the USC regime¹⁹. For a single dipole emitter in a cavity, $g \propto 1/\sqrt{V}$. However, for collective coupling, as in our system, $g \propto \sqrt{N/V}$, where N is the number of dipoles coherently coupled to the cavity mode. Thus, g depends only on the density of molecules, N/V , and is independent of the coaxial gap width (see Sec. S3).

Conclusions

We have constructed wafer-scale resonant coaxial nanocavities and achieved USC between the ENZ mode of the cavities and SiO₂ phonons with a normalized coupling strength greater than 0.25. The large magnitude of the splitting we observe – wider than the Reststrahlen band – shows that the coupling strength is truly within the USC regime, and is not the manifestation of absorption induced within the forbidden band. To further elucidate this point, we also presented a theoretical framework (both classical and quantum mechanical) that quantitatively explains the data, and which can be applied to similar USC involving homogeneous materials, waveguides, and resonant cavities.

The ENZ nanocavities can be filled with a variety of vibrationally-active materials, thus enabling USC to be realized throughout the MIR, far-IR, and terahertz spectral ranges. Furthermore, since single-nanometre-thick phononic films separate the core and cladding electrodes in each coax, the platform provides a route to combine electron tunnelling with vibrational USC and EOT, which should enable observation of ground-state electroluminescence⁴⁶ and the dynamic Casimir effect^{22,23}. The ability to reach the USC regime in mass-produced ENZ cavities with ultrasmall gap volumes ($\approx \lambda^3/10^7$) can also open up new avenues to explore quantum nonlinear optical processes⁴⁷, multiphoton effects, higher-order nonlinear effects, and single-photon excitation of multiple atoms⁴⁸, which may lead to novel applications in quantum sensing, spectroscopy, optoelectronic devices, and frequency conversion⁴⁹.

Figure captions

Figure 1. Wafer-scale resonant epsilon-near-zero (ENZ) nanocavity platform for ultrastrong coupling. (a) Geometrical parameters and illustration of SiO₂ phonons interacting with the ENZ mode of the nanocavity. (inset) Scanning electron micrograph of coaxial nanocavities with 21 nm gap, 790 nm diameter, 1190 nm period, and 80 nm Au thickness. (b) Computed field profiles (for fields polarized in the lateral direction) of a coaxial nanocavity (diameter 1120 nm and gap width 21 nm) showing a lateral section (upper) and a vertical section (lower) of the ENZ transmission resonance. (c) Process flow based on atomic layer deposition and wafer-scale glancing-angle ion milling.

Figure 2. Normal mode splittings due to ultrastrong plasmon-phonon coupling. (a) Dielectric function⁵⁰ of SiO₂ (top) and epsilon-near-zero transmission resonances (bottom) of coaxial nanoapertures (gap width of 21 nm, diameter from 700 nm to 1120 nm), simulated with a frequency-independent dielectric constant ϵ_∞ inside the aperture. This calculation shows that the bare cavity resonance sweeps through the entire Reststrahlen band for the considered diameters (Sec. S1). (b) Normalized experimental transmission spectra of SiO₂-filled coaxial Au nanocavities, for gap width of 21 nm and diameter from 550 to 1120 nm. Black dashed guidelines indicate the lower and upper polariton branches.

Figure 3. Dispersion mapping and validation of theoretical model. (a) The dispersion relation of bulk phonon polaritons supported in SiO₂ as calculated by Eq. 5 under the assumption that a single vibrational mode is excited. (b) The measured normalized coupling strength, η , and normalized mode splitting, $(2g/\gamma_{avg})$, as a function of gap size. (c-f) Dispersion maps of experimentally measured transmission spectra for coaxial apertures filled with SiO₂ with gap thicknesses of (c) 21 nm, (d) 14 nm, (e) 7 nm, and (f) 2 nm. Polaritonic resonances analytically fit using Eq. S14 (purple (upper) and yellow (lower) circle dots and solid line). ω_{ENZ} (blue circles with a solid line) is used as a fitting parameter and indicates the resonance of the ENZ mode uncoupled from the polar phonons of SiO₂. The coupling strength, g , is measured at the intersection between the uncoupled ENZ resonance and ω_{TO} (when the detuning is zero). (g) Transmission spectra measured when the bare ENZ mode crosses through the Reststrahlen band, showing distinct modal splitting and sharp resonance peaks (60% transmittance for the 21 nm-gap coax). Curves are shifted along the vertical axis for clarity.

Table 1. Mode splitting and normalized coupling strength.

Methods

Device fabrication

After pre-cleaning and pre-baking steps (200 °C for 3 min), an undoped, double-side-polished 4-inch (100) Si wafer (University wafer) is spin-coated with AZ MIR701 photoresist for 45 s at 5,000 r.p.m., followed by baking at 90 °C for 90 s. The patterns of hexagonal hole arrays with 24 different diameters from 430 nm to 1120 nm are transferred onto the Si wafer by photolithography (Canon 2500 i3 stepper) with a dose of 150 mJ/cm², followed by post-exposure bake at 110 °C and development with AZ 3000 MIF for 60 s. Hexagonal Au disk arrays are created on the Si wafer via directional evaporation of 3-nm Ti and 150-nm Au films (CHA, SEC 600), followed by liftoff using AZ Kwik Strip. After oxygen plasma cleaning (STS, 320PC) at 100 W for 30 s to remove photoresist residue, the Au disk arrays are coated conformally with a thin SiO₂ film using ALD (Cambridge Nano Tech Inc., Savannah) at a typical deposition rate of 1.2 Å/cycle. ALD was performed using ozone precursor and water vapor at 180 °C. After conformal sputtering of 3 nm Ti and 400 nm Au (AJA, ATC 2200), the top surface of 400 nm-thick Au-deposited disk patterns is planarized by glancing-angle ion milling using a 240 mA Ar⁺ ion beam incident at 5 ° from the horizontal plane (Intlvac, Nanoquest) until the top entrance of the coaxial nanocavities is exposed.

Data Availability: The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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Author contributions

D.Y. and S.-H.O. conceived the project and designed experiments. D.Y. performed device design, fabrication, and measurements. F.dL.P. and L.M.M. developed theories and performed numerical calculations. D.A.M. and I.-H.L. performed computer simulations. D.Y., F.dL.P., M.P., D.A.M., I.-H.L., M.B.R., J.D.C., L.M.M., and S.-H.O. analysed the results. All authors contributed to interpretation of the results and wrote the paper together.

Competing interests

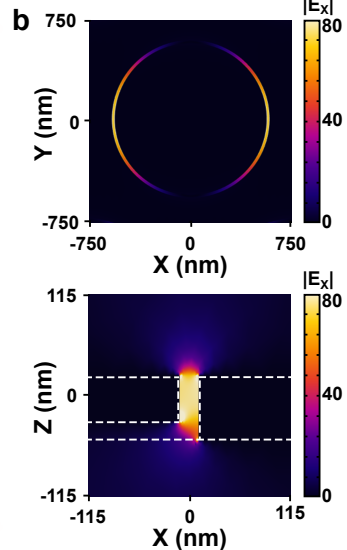
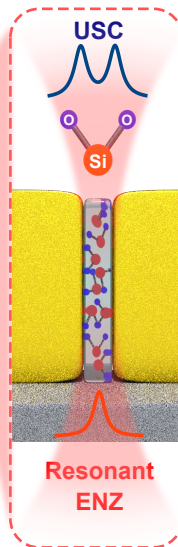
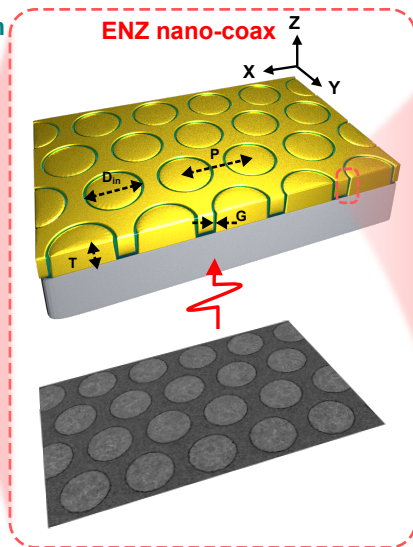
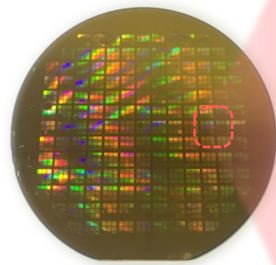
The authors declare no competing interests.

Additional information

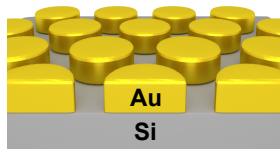
Supplementary information is available for this paper.

Correspondence and requests for materials should be addressed to S.-H.O. or L.M.M.

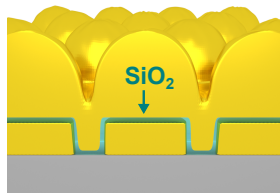
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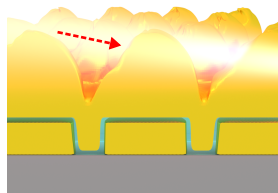
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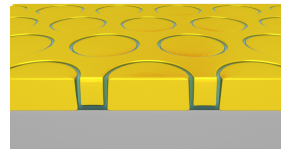
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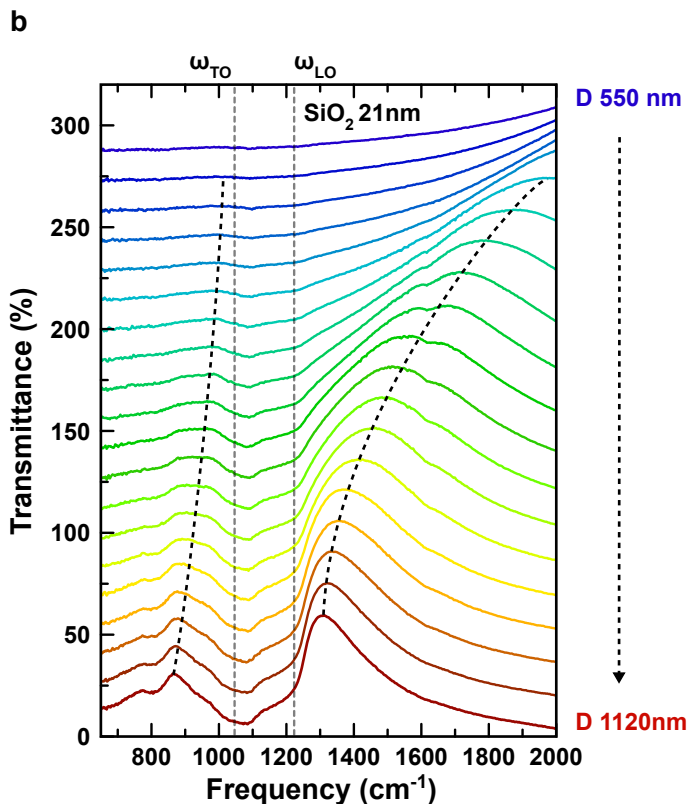
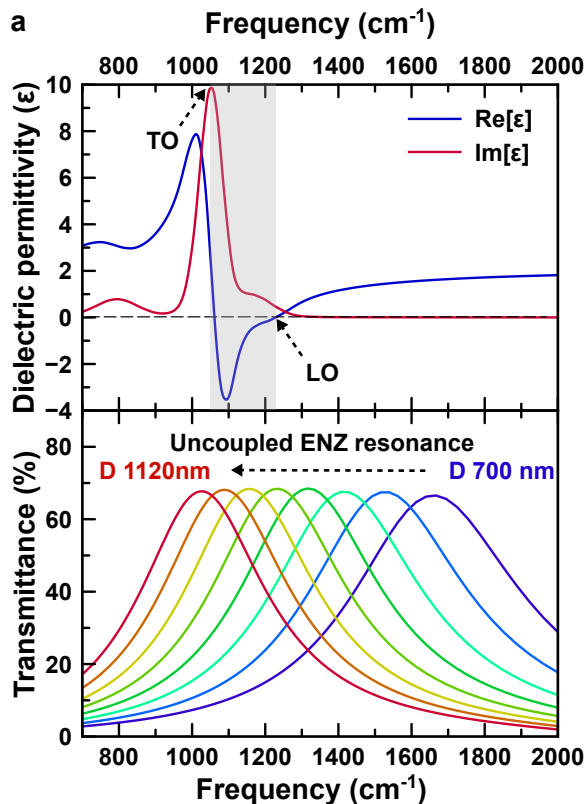


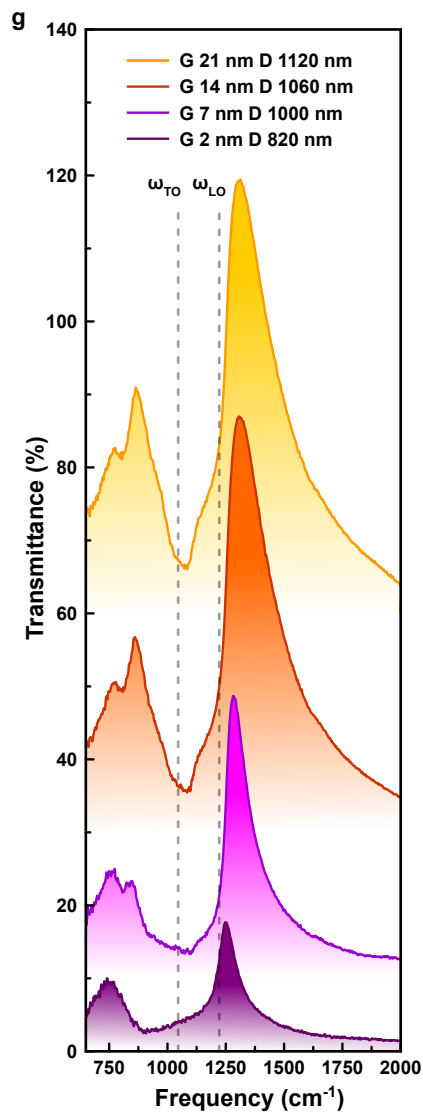
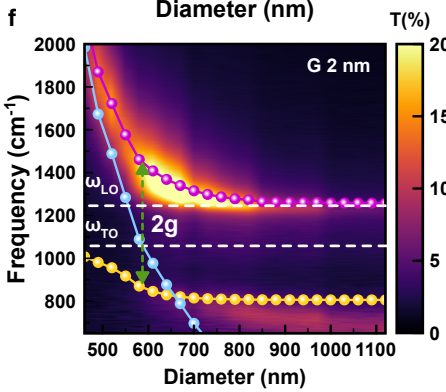
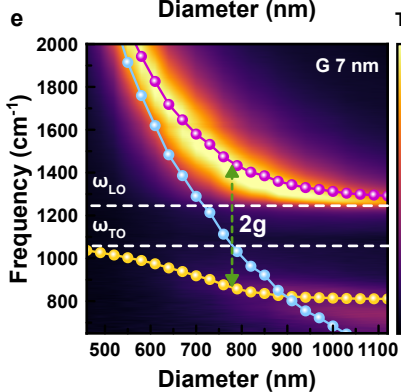
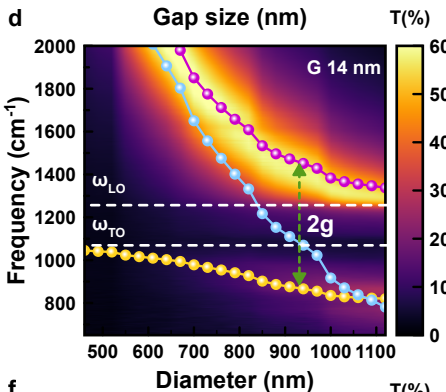
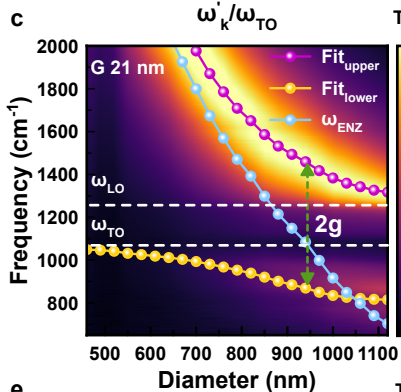
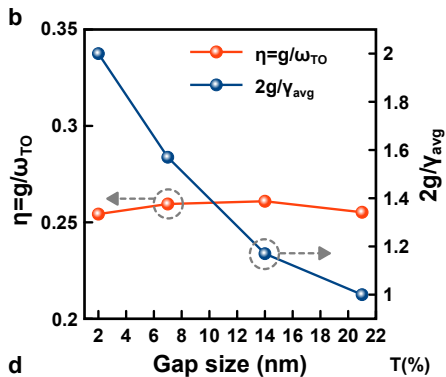
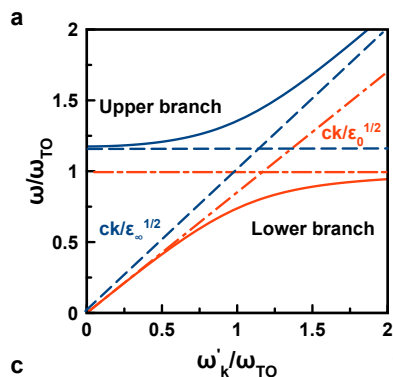
Glancing ion-milling



SiO₂-filled coaxes







SiO ₂ gap width (nm)	2	7	14	21
Bare ENZ mode linewidth (FWHM) (cm ⁻¹)	453	619	844	990
Average of bare ENZ and TO phonon linewidths, γ_{avg} (cm ⁻¹)	265	348	481	534
Normal mode splitting, $2g$ (cm ⁻¹)	534	546	549	537
Normalized mode splitting, $2g / \gamma_{\text{avg}}$	2.02	1.57	1.14	1.01
Normalized coupling strength, $\eta = g / \omega_{\text{TO}}$	0.254	0.26	0.261	0.255

Table 1. Mode splitting and normalized coupling strength.