Strong and coherent coupling between localised and propagating phonon polaritons

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Following the recent observation of localised phonon polaritons in user-defined silicon carbide nano-resonators, here we demonstrate strong and coherent coupling between those localised modes and propagating phonon polaritons bound to the surface of the nano-resonator’s substrate. In order to obtain phase-matching, the nano-resonators have been fabricated to serve the double function of hosting the localised modes, while also acting as grating for the propagating ones. The coherent coupling between long lived, optically accessible localised modes, and low-loss propagative ones, opens the way to the design and realisation of phonon-polariton based coherent circuits.

Surface phonon polaritons are surface-bound, propagative modes arising from collective oscillations of ions at the surface of polar crystals, analogous to surface plasmon polaritons on metallic surfaces [1, 2]. When the surface is properly patterned, it can sustain also localised surface phonon polaritons, confined in extremely subwavelength volumes and characterised by quality factors and Purcell enhancements unparalleled in plasmonic systems [3, 4]. Patterning, apart from creating the localised modes, also acts as a grating for phase-matching to propagating surface polaritons [5], allowing to tune their dispersion, and making it possible to bring them in resonance with the localised ones. The possibility to couple long lived localised resonances that can be resonantly pumped by an external source [5] to low-loss propagative modes, with propagation lengths of hundreds of micrometers [6], hints to the tantalising prospect to observe quantum effects in those systems, analogously to what was done with surface plasmon polaritons [7–9]. Moreover, the unique properties of phonon polariton resonators could lead to the realization of phonon-polariton based quantum circuits, overcoming the main problems hampering the development of quantum plasmonic circuits [10]. Here, using a silicon carbide (SiC) surface patterned by micrometer-sized cylinders, we demonstrate strong coupling between localised and surface modes by presenting clear evidence of spectral anticrossing, thus implying a coherent, reversible energy exchange [11, 12]. Our work thus validates different building blocks toward a novel technological platform for coherent mid-infrared applications.

How tightly light of a given frequency may be confined is limited by the bandwidth of spatial frequencies available. The most famous example of this is the Abbe diffraction limit but the phenomenon is pervasive. Piecewise homogeneous material systems can sustain electromagnetic resonances localised around interfaces where the permittivity changes sign, the out-of-plane wavevector becoming imaginary and the bandwidth of spatial frequencies in-plane increasing. For a flat surface between air and a material with negative permittivity $\epsilon(\omega)$, this leads to surface modes characterised by the well known dispersion

$$q = \frac{\omega}{c} \sqrt{\frac{\epsilon(\omega)}{\epsilon(\omega) + 1}}$$

(1)

where $q$ is the in-plane wavevector and $c$ is the speed of light. Surface plasmon polaritons are well known surface modes in metals, whose Drude permittivity $\epsilon_D(\omega) = 1 - \frac{\omega_p^2}{\omega^2}$ becomes negative due to the coupling with collective plasma excitations in the region below the plasma frequency $\omega_p$ [13]. Strong field localisations are achievable in plasmonic systems, with good applications in waveguiding [14] and usually inefficient processes such as Raman spectroscopy [15]. Still, plasmons are inherently lossy [16], the modal energy spending half cycles as electron kinetic energy, leading to a dominant loss channel of electron-electron scattering occurring on the 0.01-ps scale [17], thus making it challenging to integrate them in quantum technology architectures [10, 18]. As an alternative platform to metals, also polar dielectrics support surface polaritons in-between the frequencies of the transverse optical phonon, $\omega_{\text{TO}}$, and the longitudinal optical phonon, $\omega_{\text{LO}}$, where the Lorentz permittivity $\epsilon_L(\omega) = \frac{\omega_p^2 - \omega_{\text{TO}}^2}{\omega^2 - \omega_{\text{TO}}^2}$ becomes negative as a result of light coupling to oscillations of the ions [19]. The damping of the ionic oscillations occurs on the 1-ns scale, two orders of magnitude slower than electron damping in metals. The resulting modes, called surface phonon polaritons [1], have been exploited for a number of applications, from enhanced energy transfer [20] and waveguiding [21] to thermal coherent infrared emission [22, 23], super-lensing [24, 25], near field optics [26], enhanced optical forcing [27], and sensing [28]. More exotic physics is expected in recently observed hyperbolic materials such as hexagonal boron nitride [29]. Analogously to localised plasmonic resonances, localised phonon resonances also appear in subwavelength dielectric systems. Mutschke [30] carried out explicit investigations into the infrared properties of small SiC particles of various polytype observing morphology dependent resonances analogous to particle plasmons. Subwavelength SiC whiskers have also been shown to support both localised electrostatic...
FIG. 1: a) Fundamental dispersion of the surface mode is given by the black curve. Solid blue curves indicate surface mode folding from the edge of the first Brillouin zone (indicated by corresponding vertical dashed lines) for periodicities 5µm-7µm. The red curve shows the vacuum light line. The inset shows the electric field norm for a surface mode at an air / SiC interface. The grey shaded region in the inset shows the beginning of the Si substrate on which the SiC wafer we used is grown, highlighting that the surface mode is almost entirely localised into the SiC region. b) Tight-binding dispersion of the monopolar mode of a pillar array is indicated by blue curves for a variety of periodicities. The red curve is the vacuum light line, with c the speed of light. Insets show a slice of the mode electric field norm in an isolated SiC cylinder on substrate, calculated using COMSOL Multiphysics, and a SEM image of a fabricated resonator. c) SEM image of the fabricated array. d) Electric field norm for a mode of the coupled array. The sinusoid indicates the surface mode wavelength.

and propagative Fabry-Perot modes [3]. Recently advances in fabrication procedures have allowed for the creation of user-defined cylindrical SiC nano-resonators on SiC substrate [4, 31]. The modes exhibit quality factors exceeding the theoretical limit for plasmonic resonators. Moreover, while absolute confinement of light is less than in plasmonic systems due to the longer wavelengths involved, reducing potential nonlinear effects, relative confinement (in units of the wavelength) is comparable with the better plasmonic resonators [32], leading to Purcell enhancements 4 orders of magnitude greater than comparable plasmonic systems [4].

We used a 9.7µm thick planar 3C-SiC layer grown over Si substrate, on which subwavelength cylindrical resonators of height ≃ 800nm and diameter ≃ 1µm were fabricated by ICP RIE in square 70 × 70 pillar arrays of varying periods from 5µm to 7µm. Numerical calculations show this depth of SiC is sufficient for convergence of reflectance measurements to those of bulk SiC, as can also be inferred from the inset in Fig. 1(a) where we see that the electric field of the surface mode in the dielectric is completely segregated into the SiC region. Full details on the fabrication can be found in the Supplemental Material. The planar surface supports a surface mode whose dispersion, ω_q, is given by Eq. (1) with ϵ(ω) the dispersive SiC dielectric function. While the surface polariton dispersion usually lies outside the lightcone, the periodic patterning of the substrate results in a period dependant band folding of the dispersion at the edge of the first Brillouin zone, as illustrated in Fig. 1(a), thus making them optically accessible. In the inset the electric field norm of a surface mode is plotted. The isolated cylinder-on-substrate system supports a number of modes as dis-
The objective illuminates directionally and the sample is reflectance mode utilising a grazing incidence objective. Fabricated arrays were measured by FTIR microscopy in an anticrossing in the dispersion of the normal modes. Coupling between localised and surface modes leads to dashed lines in Fig. 2(a). In order to highlight the subwavelength character, the normal modes, from Eq. (2), can be diagonalised by a Hopfield-Bogoliubov procedure [36] in terms of two free normal modes, whose annihilation operators read

\[ \hat{p}_q^+ = X_q \hat{a}_q + Y_q \hat{b}_q, \quad \hat{p}_q^- = Y_q \hat{a}_q - X_q \hat{b}_q \]  

where \( X_q \) and \( Y_q \) are the Hopfield coefficients describing the mixing of surface and localised modes and the frequency of the normal modes is

\[ \omega_q \pm = \sqrt{\omega_q^m + \omega_q^p \pm \sqrt{(\omega_q^m - \omega_q^p)^2 + 4g_0^2}}. \]  

The simulated electric field norm for a mode of the coupled system is shown in Fig. 1(d), where for comparison, and in order to highlight the subwavelength character of the coupling, we explicitly show a typical wavelength for the resonant surface mode. A typical dispersion of the normal modes, from Eq. (4), is shown by the dot-dashed lines in Fig. 2(a), in which it is clear how the coupling between localised and surface modes leads to an anticrossing in the dispersion of the normal modes. Fabricated arrays were measured by FTIR microscopy in reflectance mode utilising a grazing incidence objective. The objective illuminates directionally and the sample is aligned so the peak incident intensity is along the principal axis of the resonator array. Details of the measurement are given in the Supplemental Material. High-angle illumination is achieved by use of a mirror to rotate the incident beam onto the sample resulting in a dual peaked angular excitation as illustrated in Fig. 2(b). This allows two slices of the polariton dispersion to be measured simultaneously as shown in Fig. 2(a). Note that we have until now neglected losses in our theoretical treatment, on account of the large quality factors of both localised

\[ \mathcal{H} = \sum_q \hbar \omega_q^m \hat{a}_q \hat{a}_q + \hbar \omega_q^p \hat{b}_q \hat{b}_q + \hbar g_0 \left( \hat{a}_q \hat{b}_q + \hat{a}_q \hat{b}_q^\dagger \right) \]  

where \( \hat{a}_q \) and \( \hat{b}_q \) are the bosonic creation operators for the monopolar modes and surface modes respectively. As detailed in the Supplemental Material, the Hamiltonian in Eq. (2) can be diagonalised by a Hopfield-Bogoliubov procedure [36] in terms of two free normal modes, whose annihilation operators read

where \( X_q \) and \( Y_q \) are the Hopfield coefficients describing the mixing of surface and localised modes and the frequency of the normal modes is

\[ X_q = \frac{\omega_q^m + \omega_q^p - \sqrt{(\omega_q^m - \omega_q^p)^2 + 4g_0^2}}{2}, \quad Y_q = \frac{\omega_q^m + \omega_q^p + \sqrt{(\omega_q^m - \omega_q^p)^2 + 4g_0^2}}{2}. \]  

FIG. 2: a) Dispersion for array period 6.25\( \mu \)m and coupling constant \( g_0 = 1.63\) meV (13.1/cm). Purple dotted lines are the constituent monopolar and folded surface phonon polariton branch, coupled normal modes are green dash-dotted lines. Blue dashed lines represent the two different angles \( \theta_1 \) and \( \theta_2 \) sampled by the dual illumination. Squares and circles are the fitted data points from the reflectance plots in the Supplemental Material. b) Illustration of the function of the grazing incidence objective. A Schwarzschild light path is indicated by angularly symmetric blue rays. The plane mirror at the bottom breaks the cylindrical symmetry of the objective rotating the light cone, initially at an incidence angle \( \theta \), toward the sample. After reflecting upon the sample surface the beams strike a spherical mirror and are focussed on the same spot before passing back into the objective. This results in a dual non-normal double-pass illumination at angles \( \theta_1 \) and \( \theta_2 \). Notice that the separation between the two rays is exaggerated here for clarity purpose. The actual data for both the rays’ incidence angles and their angular spread can be found in the Supplemental Material.
reflectance map we obtained is given in Fig. 3(a) as a function of the array period, that is tuning the surface mode resonance. The data exhibits a clear anticrossing when the two modes are resonant, showing that the system is indeed in the strong coupling regime. Peak positions were then extracted from the experimental reflectance map, clearly highlighting the presence of peaks from two different angles, not apparent in Fig. 3(a) due to the small angular shift and finite linewidth. Individual spectra measured for each array period, and the relative calculated fits can be found in the Supplemental Materials. The data was then fitted, following the procedure detailed in the Supplemental Material, to the normal mode dispersion given in Eq. 4. The phenomenological coupling $g_0$ was assumed to vary super-radiantly with the in-plane resonator density analogously to scalings in systems where surface plasmons interact with molecular excitons, where in our case the resonators act as effective molecules [37]. The peak positions extracted from the experimental data and the resulting fits are given in Fig. 3(b), where we explicitly show the dispersions at the two different angles. The model reproduces well the anticrossing, within errors of the order of 1 meV (8/cm), a similar magnitude to those reported in previous simulations using finite element modelling [4]. Those errors have been attributed to modifications of the dielectric properties of SiC near the surface due to the strain induced in SiC grown on Si substrates due to the mismatch of lattice parameters [38]. In order to verify this hypothesis we repeated the fitting procedure using the high and low frequency values of the dielectric constants and the TO phonon frequency as additional fit parameters. The resulting values for the dielectric parameters differ less than 5% from the standard values found in the literature [19] and the TO phonon shifts just 0.74 meV (6.1/cm), but they lead to a dramatic improvement of the fits, shown in Fig. 3(c). The maximal value of the fitted Rabi frequency is $g_0 = 2.55$ meV (20.6/cm), leading to a ratio between $g_0$ and the bare frequency of the excitation of the order of $10^{-2}$, thus justifying a posteriori the rotating wave approximation we used in Eq. (2) [39]. We also extracted the linewidths of the different normal modes from the reflectance map, and we were able to fit them assuming they are sums of the constituents’ ones, weighted by the relative Hopfield coefficients [40]. We found linewidths at the anticrossing of the order of 1 meV (8/cm). In the densest array we considered, energy is thus coherently transferred between monopolar and surface modes roughly 4 times before escaping. More details, including the extracted experimental linewidths, can be found in the Supplemental Material.

In summary, we have demonstrated that, exploiting the tunability of surface phonon polaritons dispersion, it is possible to observe a clear spectral anticrossing between localised and surface phonon polaritons, proving that coherent, reversible energy exchange is possible between them. In combination with the high con-

FIG. 3: The top panel (a) shows the background subtracted experimental reflectance map of SiC cylinder arrays of varying period. The almost dispersionless mode at 113.75 meV (917.4/cm) is the transverse dipole resonance discussed elsewhere [4]. The peaks extracted from the reflectance map are given in the lower panels for the larger angle by blue squares and the smaller by red circles. Solid blue lines and dashed red lines are the corresponding fits, enacted using book values for the dielectric constants of SiC (b) [19], or fitting also the dielectric constants of SiC (c) as free parameters.

and surface phonon polaritons. Still, it is important to remember that the anticrossing shown in Fig. 2(a) is present only if the Rabi frequency $g_0$ is larger than the losses of both modes, including pure dephasing [11], a condition usually referred to as strong coupling regime. Observing an anticrossing in the system spectrum thus unequivocally proves that energy can be reversibly and coherently exchanged between the two modes, fulfilling the main requirements to use them as building blocks for coherent circuit architectures [12]. The experimental reflectance map we obtained is given in Fig. 3(a) as a
finements and Purcell enhancements recently observed in user-defined structures, the present Letter takes a decisive step in demonstrating the versatility and tunability of phonon polaritons for coherent applications in the mid-infrared spectral region. In particular, the coherent interplay between localised and propagative, nonradiative modes, together with the relatively large quality factors achievable in those systems, could make it possible to design quantum architectures similar to quantum plasmonic circuits, but without many of the limitations due to plasmonic intrinsic losses.

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Fabrication

The surface phonon polariton resonators were fabricated starting from a polished 9.7\mu m thick layer of <100> oriented 3C-SiC, grown heteroepitaxially on a <100> Si substrate (NOVASIC). Patterning was carried out via standard liftoff process, using a bilayer of MMA-PMMA (thickness 250nm and 150nm respectively) exposed by electron beam lithography. The bilayer allowed a deposition of 120nm of Ni hardmask via electron beam evaporation. The sample was kept in acetone until the unpatterned Ni was completely removed and subsequently dry etched via ICP RIE in a SF6 and Ar chemistry at 0.7mTorr pressure, 280W of bias power and 800W of ICP power. Finally, the Ni hardmask was removed with a fuming nitric acid wet etch for 20\,'. The dry etch was calibrated to obtain 811\pm8nm high structures and shows an etching angle of 86.5 degrees.

Measurements

The reflectance map in Fig. 3(a) of the main text was recorded using a Bruker Hyperion 2000 microscope. The standard internal MIR source of a Bruker Vertex 70 FTIR was utilised to illuminate over the entire spectral range simultaneously. The sample was illuminated under TM polarisation with use of a grazing incidence objective which utilises a mirror to rotate the incident light cone directionally onto the sample. This means that the sample is effectively measured at two different incidence angles at the same time, resulting in each resonance being split into two narrowly separated peaks, as illustrated in Fig. 2(b) of the main text. Knife edge apertures were utilised to restrict collected data to the geometrical area of the pillar array. The reflectance from the sample was measured utilising a cooled MCT detector with spectral resolution of 0.25/cm. Background subtracted reflectance plots for each array period are shown in Fig. 1, experimental points are indicated by circles and the solid cyan line is the numerical fit. The data was fitted with a multiple Lorentzian fit for all peaks recorded, comprising the modes of interest and other background modes at higher energies within the Restrahlen band. The four resonances utilised in the fitting procedure, corresponding to the two normal modes due to the strong coupling between localised and propagative phonon polariton modes, each one at two different angles, are indicated by dashed red lines in the figure. Other resonances at higher energies, indicated by dash-dotted green lines, correspond to the higher excited states of the resonators. The first resonance in particular, close to 920/cm and clearly visible in Fig. 3(a) of the main text, corresponds to the first transverse mode of the pillars, already observed in Ref. [4]. These higher-lying resonances are not utilised in the polariton fitting procedure described in the main text.

Theoretical Modelling

The dispersion of the uncoupled cylinder array is described by a tight binding model as

\[
\omega^m_q = \omega^c \left( 1 + \frac{1 + \sum_{n \neq 0} e^{-inqR} \beta_n}{1 + \Delta \alpha + \sum_{n \neq 0} e^{-inqR} \alpha_n} \right)
\] (S1)

where \( n \) indicates a discrete resonator, \( \alpha_n, \beta_n \) and \( \Delta \alpha \) are overlap integrals as defined in Ref. [1], \( R \) is the array period, and \( \omega^c \) is the frequency of the monopolar mode of a single cylinder. We consider only nearest neighbour interactions in the relevant limit where the coupling between resonators is small due to the large separations, and the tight-binding equation simplifies to

\[
\omega^m_q = \omega^c \left( 1 - \frac{\Delta \alpha}{2} + \kappa_1 \cos(qR) \right)
\] (S2)
FIG. 1: Experimental background subtracted reflectance data is given by the circles for array periods a) 5µm, b) 5.5µm, c) 5.75µm, d) 6µm, e) 6.25µm, f) 6.5µm and g) 7µm. The data is fitted by a multiple Lorentzian fit indicated by the solid blue line. Other lines are the individual Lorentzian curves comprising the fit. Dashed red lines are those utilised for the polariton model, while the dash-dotted green ones correspond to higher-energies localised modes.
where \( \kappa_1 = \beta_1 - \alpha_1 \). The remaining tight-binding parameters \( \kappa_i \) and \( \Delta \alpha \) are assumed to have a dipole-dipole like dependency on the resonator separation, modelled as \( 1/R^3 \). The appropriateness of this model was independently confirmed by numerical simulations carried out in the RF module of COMSOL Multiphysics. As the surface mode wavelength is larger than the pillars separation, the surface-monopole coupling strength \( g_0 \) is assumed to vary super-radiantly with the in-plane resonator density \( \rho \) as \( \sqrt{\rho} \). The appropriateness of this model was independently reported scalings in systems consisting of surface plasmons interacting with molecular excitons, where in our case the resonators act as effective molecules as in Ref. [2].

The Hamiltonian

\[
\mathcal{H} = \sum_q \hbar \omega_q^m \hat{a}_q^\dagger \hat{a}_q + \hbar \omega_q^s \hat{b}_q^\dagger \hat{b}_q + \hbar g_0 \left( \hat{a}_q^\dagger \hat{b}_q + \hat{b}_q^\dagger \hat{a}_q \right)
\]

(S3)

can be put in diagonal form by diagonalising the corresponding Hopfield-Bogoliubov matrix for each value of the in-plane wavevector \( q \)

\[
H_q = \begin{bmatrix} \omega_q^m & g_0 \\ g_0 & \omega_q^s \end{bmatrix}
\]

(S4)

whose eigenvalues are

\[
\omega_q^\pm = \frac{\omega_q^m + \omega_q^s \pm \sqrt{(\omega_q^m - \omega_q^s)^2 + 4g_0^2}}{2}
\]

(S5)

and the respective eigenvectors \([X_q, Y_q]\) give the Hopfield coefficients appearing in the expression of the polaritonic operators

\[
\hat{p}_q^+ = X_q \hat{a}_q + Y_q \hat{b}_q \quad \text{(S6)}
\]

\[
\hat{p}_q^- = Y_q \hat{a}_q - X_q \hat{b}_q.
\]

This system may also be probed numerically. An angled resolved reflectance map calculated utilising the RF module of the commercial finite element software COMSOL Multiphysics for a SiC cylinder array of period 6 \( \mu \)m composed of cylinders of height 800 \( \text{nm} \) and diameter 1 \( \mu \)m on semi-infinite SiC substrate is shown in Fig. 2. The same anticrossing behaviour is observed between the longitudinal cylinder mode and the mode of the planar SiC substrate, confirming the experimental results.

**Fitting**

In order to fit the spectrum of the system as a function of the array period, as shown in Fig. 3(b) of the main text, least squares fits were carried out for the tight binding parameters \( \kappa_1, \Delta \alpha, \omega^s \), the two incident angles \( \theta_1, \theta_2 \) shown in Fig. 2(a), and the parameter \( \zeta \), linked to the coupling strength as \( g_0 = \zeta \sqrt{\rho} \). For Fig. 3(c) also the high and low frequency dielectric constants \( \epsilon_\infty \) and the TO phonon frequency \( \omega_{TO} \) were used as fitting parameters. The fitting procedure yielded \( \theta_1 = (48.54 \pm 0.05)^\circ \), \( \theta_2 = (55.08 \pm 0.08)^\circ \), \( \epsilon_\infty = 9.26 \pm 0.22 \), \( \epsilon_\infty = 6.68 \pm 0.17 \) and \( \omega_{TO} = 97.85 \pm 0.07 \text{meV} \) (789.3 \pm 0.6/cm).

**Linewidths**

We extracted the linewidths of the different normal modes from the reflectance map. In order to model them we assumed they are sums of the constituents’ ones, weighted by the relative Hopfield coefficients [3]

\[
\gamma_q^+ = \gamma_m |X_q|^2 + \gamma_s |Y_q|^2
\]

(S7)

\[
\gamma_q^- = \gamma_m |Y_q|^2 + \gamma_s |X_q|^2
\]

where \( \gamma_m \) is the linewidth of the monopolar mode, essentially constant over the measured region, and \( \gamma_q^+ \) is the dispersive linewidth of the surface mode. The surface phonon polariton mode results from strong coupling of photons to the transverse phonon resonances of the crystal. It can thus be described in the same Hopfield-Bogoliubov framework we
used previously as a linear superposition of free photons and optical phonons components, whose Hopfield coefficients we denote $C_q$ and $D_q$ respectively. As the free photon dispersion is very steep, those coefficients depend strongly on the wavevector. The linewidth of the surface mode therefore also obeys the equivalent of Eq. (S7),

$$
\gamma^s_q = \gamma^{\text{ph}}|C_q|^2 + \gamma^{\text{TO}}|D_q|^2
$$

(S8)

where $\gamma^{\text{ph}}$ and $\gamma^{\text{TO}}$ are respectively the loss rates of the photonic and phononic components. Given the very small value of the intrinsic photonic losses, mainly due to surface roughness or spurious absorption we can safely assume $\gamma^{\text{ph}} = 0$. In addition to the previously considered intrinsic broadening, the measured resonances are further broadened due to the finite angular aperture of the objective, leading to an extra broadening in first approximation proportional to the slope of the mode dispersion. The extra dispersive broadening to be added to the intrinsic one is thus given by the expression

$$
\gamma^{\text{ds}}_q = \frac{\partial \omega^{-\pm}_q}{\partial \theta} \Delta \theta = \frac{\partial \omega^{-\pm}_q}{\partial q} \frac{\partial q}{\partial \theta} \Delta \theta = \frac{\omega^{-\pm}_q v^{-\pm}_q}{c} \cos \theta \Delta \theta,
$$

(S9)

where $\theta$ is the peak illumination angle along $q$, $v^{-\pm}_q$ is the group velocity of the polariton mode, and $\Delta \theta$ the angular aperture.

The model was parametrized using the Hopfield coefficients fixed by the fitting procedure for the spectrum, and then fitted to the measured linewidths with Eq. (S7) and Eq. (S8), using $\gamma^{\text{TO}}$, $\gamma^m$, and $\Delta \theta$ as fitting parameters. Experimental data and the fits are given in Fig. 3. The lower polariton is seen to broaden dramatically at large pitches due to the strong dispersion and finite angular spread sampled. From the fitting procedure we obtain $\gamma^{\text{TO}} = 4.1$/cm, $\gamma^m = 10.6$/cm, and $\Delta \theta = 0.1$ rad. The predominant effect in this system is the dispersive broadening. The calculated Q-factor for the monopolar mode is 81 which falls in the centre of the range previously measured [4]. The book value of $\gamma^{\text{TO}}$ based on a single oscillator fit to the experimental dielectric function is 6/cm [5], similar variations in the
parameters $\omega^{\text{TO}}$ were observed on fitting for the energies as discussed in the main text. The value of $\Delta \theta = 0.1$ rad corresponds to an acceptance angle of around 5.7° for each illumination beam.

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