

Strong coupling between a dielectric nanocavity and a monolayer transition metal dichalcogenide

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We demonstrate strong coupling between light in a dielectric nanocavity with deep subwavelength confinement and excitons in a monolayer of molybdenum ditelluride. Avoided crossing is demonstrated by both photoluminescence and reflection measurements, from which we extract a light-matter interaction strength of $g_{\text{PL}} = 5.3 \pm 0.3$ meV and $g_{\text{R}} = 4.7 \pm 0.7$ meV, respectively. The associated Rabi splitting is twice as large as the system's losses. These values are in good agreement with values obtained by an exciton reaction coordinate formalism, yielding $g_{\text{theory}} = 5.2 \pm 0.7$ meV. The strong light-matter interaction, combined with low losses and subwavelength confinement of light, demonstrates a regime of light-matter interactions where strong nonlinearities at the single-photon level are expected.

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Monolayer (ML) transition-metal dichalcogenides (TMDCs), consisting of one molybdenum atom and two chalcogen atoms per unit cell, are direct band gap semiconductors in the 2H phase [1,2]. Their unique excitons, featuring high binding energy [3,4], high absorption coefficient [5,6], and large oscillator strength [7,8], make them compelling platforms for exploring strong light-matter interactions [9]. Strong coupling occurs when the energy exchange between light and excitons is fast enough to surpass the system's decay rate, resulting in Rabi oscillations and hybridized polariton states [10–12]. The regime of strong light-matter interaction has been demonstrated with excitons in ML TMDCs coupled to optical microcavities, where light is confined on length scales similar to or larger than the wavelength of the light [13–18]. As these structures are based on dielectric materials, cavity-induced losses are small and cavity linewidths are typically on the order of 0.1–10 meV. Other ways to achieve strong light-matter interactions with TMDCs and periodically patterned dielectric structures are bound states in the continuum [19–21] and metasurfaces [22,23]. Similarly, TMDCs integrated with plasmonic structures, such as surface plasmon polaritons [24,25] and plasmonic nanocavities [26–29], can exhibit strong light-matter interactions. In the latter case, light is confined on deep subwavelength length scales, but the confinement is accompanied by significant metal-induced losses and plasmonic linewidths typically exceed 100 meV.

Integrating dielectric cavities, such as photonic crystal cavities [30,31] and nanobeam cavities [32], with ML molybdenum ditelluride (MoTe_2) is particularly promising due to its excitonic transition energy in the near-infrared [33]. This has led, for example, to weak coupling and enhanced spontaneous emission [34]. While recent advances have explored the light-matter interactions of ML TMDCs with dielectric nanobeam cavities [35,36], strong coupling in these systems has not yet been achieved.

Recently, confinement of light on subwavelength scales using dielectric materials was realized [37–43]. Inverse design by topology optimization has been demonstrated as a versatile tool for designing these cavities with extreme dielectric confinement (EDC) [44–46], and topology-optimized EDC cavities have been experimentally realized in silicon [42] and indium phosphide (InP) [43]. The subwavelength confinement, which was previously achievable only with plasmonic structures [47–49], comes without the associated ohmic and absorption losses intrinsic to plasmonic materials and therefore enables intriguing possibilities [50–53]. In particular, a recent study demonstrated lasing from a quantum well embedded in such an EDC cavity [54].

Similarly, the integration of EDC cavities with excitons in monolayer TMDCs unlocks possibilities for exciton dynamics and device applications. Exciton-polaritons confined by a cavity mode to a single, subwavelength-scale region have been shown to exhibit strong exciton-exciton interactions [57–59]. The combination of near-plasmonic subwavelength confinement, low-loss dielectric cavities, and the strong oscillator strength of excitons in monolayer TMDCs enables a regime for exploring strong light-matter interactions. Studying polaritons in this regime is of significant interest, as a nonlinearity in an interacting system yields photon antibunching [60,61]. In particular, recent theoretical work [62] has shown that enhanced quantum nonlinearities at the single-photon level could be achieved, enabling a polariton blockade [62–64].

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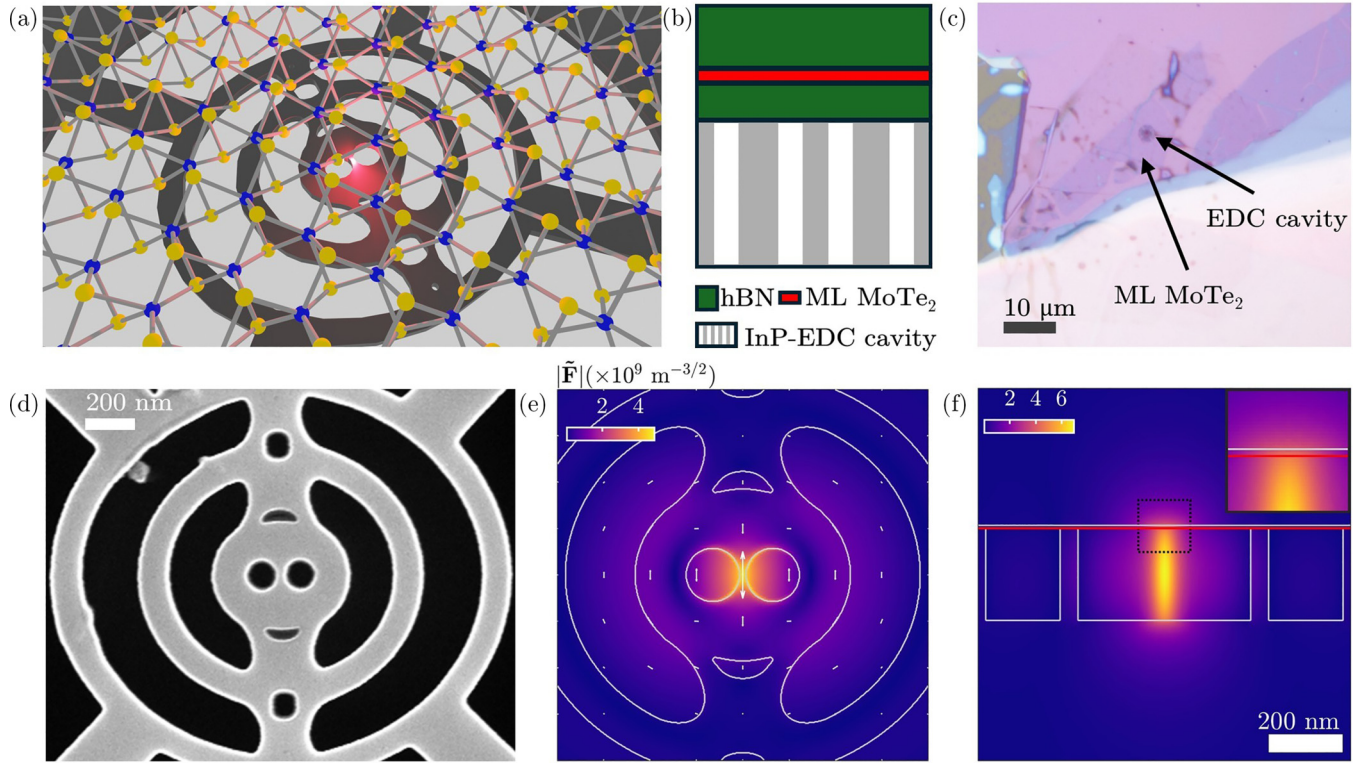


FIG. 1. (a) Artistic representation of the system. Light is confined in an EDC cavity. The ML MoTe₂ lies on top of the EDC cavity [55,56]. The hBN flakes are not depicted. (b) Sketch of the side view of the structure. (c) Microscope image of the hBN-encapsulated ML MoTe₂ on the EDC cavity. (d) Scanning electron microscope (SEM) image of the passive cavity. Simulated electric-field distribution of the structure in (e) the xy plane at $z = z_{2D}$ and (f) the yz plane at $x = 0$. Contour lines show the topography of the cavity and the hBN layer. Arrows in panel (e) depict the in-plane polarization of the electricfield. The red line in panel (f) depicts the ML MoTe₂.

Moreover, the spin-valley selection rules of ML TMDCs support the study of chiral light-matter interactions [65–68], while the system also serves as a promising platform for exploring condensation effects [50]. Unlike plasmonic nanostructures, dielectric cavities integrate seamlessly with existing technology [41,52,69,70], offering significant scalability and application benefits.

In this work, we experimentally demonstrate strong coupling between an InP-based EDC cavity with subwavelength confinement and excitons in ML MoTe₂. The avoided crossing of the system is demonstrated with temperature-resolved photoluminescence (PL) and reflection measurements, in which the excitonic transition and the cavity mode are resonant at $T = 40$ K. The extracted light-matter interaction strength is approximately 5 meV. The extracted cavity linewidth is 3.3 ± 0.1 meV, more than an order of magnitude smaller than typical plasmonic linewidths, while a lateral field confinement length of $\sigma \approx 70$ nm is maintained, more than a factor of 2 smaller than $\lambda/(2n)$. The obtained Rabi splitting is twice as large as the losses of the system, and the experimentally obtained value is in agreement with numerical simulations based on the reaction-coordinate formalism [59]. This marks an important step toward the observation of exciton-exciton interactions and single-photon nonlinearities [62] in integrable photonic devices operating close to the O band.

An artistic representation of the ML MoTe₂ on an EDC cavity is depicted in Fig. 1(a). ML MoTe₂ is sandwiched in

hexagonal boron nitride (hBN) to prevent degradation of the monolayer [71] and to reduce the linewidth [34,72–74]. The resulting hBN/MoTe₂/hBN heterostructure is placed on the EDC cavity [cf. Fig. 1(b)]. The MoTe₂ layer is separated by 1.5 ± 1.3 nm from the surface of the structured InP, and we refer to the z position of the ML MoTe₂ as z_{2D} . See Sec. S1 in the Supplemental Material [75] for details on the two-dimensional (2D) material fabrication process. A microscope image of the resulting sample is depicted in Fig. 1(c). Due to the large extent of the ML MoTe₂ compared to the cavity mode, the excitons are considered delocalized in the plane but strictly confined in the z direction [59]. Nevertheless, the exciton-polaritons are laterally confined due to interaction with the subwavelength cavity mode.

The EDC cavity design results from a simplified version of a topology-optimized (TO) structure, where the TO structure is approximated by only ellipses and tangents. This smoothes the topography while maintaining the overall structure of the original TO design, which has been shown to successfully retain the properties of the cavity [76]. A scanning electron microscope image of a similar cavity on the same chip can be found in Fig. 1(d). The outer rings of the cavity increase the Q factor by suppressing radiative losses, similar to circular Bragg reflectors, while the holes in the center provide extreme dielectric confinement on subwavelength scales. The confinement is largely determined by the spacing of the void regions in the cavity center. A void spacing of 20 nm was targeted, safely above what has been determined as a

fabrication limitation [43] while yielding a subwavelength field confinement (see below). The previous design targeted a resonance in the telecom *C* band. To acquire a design where the cavity resonance energy matches the excitonic transition of the A exciton in ML MoTe₂ at cryogenic temperatures, the design was therefore scaled appropriately. The EDC cavity is fabricated in a 245 nm thick InP membrane [43]. This material system is chosen because the excitonic transition in MoTe₂ is below the band gap of InP. Therefore, the system is not limited by absorption losses of the dielectric material.

The dominant response of the optical nanocavity can be accurately described by a single quasinormal mode (QNM) [77–82] with complex eigenenergy of the form $\tilde{E}_{\text{cav}} = E_{\text{cav}} - i\Gamma_{\text{cav}}/2$, in which E_{cav} denotes center position and Γ_{cav} is the full width at half maximum of the emission spectrum in the single-mode approximation. From numerical calculations, as detailed in the Supplemental Material [75], we find that the structure supports a QNM with nominal resonance energy of $E_{\text{cav}} = 1.187$ eV, a nominal quality Q factor of $Q = 571(7)$, and an effective mode volume of $V_{\text{eff}} = 0.060(2)(\lambda/n)^3$. We note, however, that dielectric nanocavities often exhibit significantly lower Q factors than predicted by simulations [42,43], and we extract an experimental Q factor of 358(11); see Sec. S1 in the Supplemental Material [75]. This reduction is primarily attributed to fabrication imperfections, such as surface roughness and geometric deviations [83]. Owing to their extreme field confinement, EDC cavities exhibit pronounced sensitivity to such imperfections [43]. Moreover, the resonance frequency is highly sensitive to small variations of the geometry [76]. Figures 1(e) and 1(f) show the magnitude of the normalized field profile $|\tilde{\mathbf{F}}(\mathbf{r})|$ in the *xy* plane at z_{2D} and in the *yz* plane at $x = 0$, respectively. The arrows in Fig. 1(e) depict $\Re[F_{x,y}(\mathbf{r})]$, highlighting the linear polarization of the cavity $\hat{E}_{\text{cav}}$ along the *y* axis [76,84]. The lateral confinement length of the field in the MoTe₂ plane is estimated as $\sigma \approx 70$ nm; see Sec. S2 in the Supplemental Material [75]. The structure also supports another mode with orthogonal polarization, lower resonance energy, and reduced quality factor—rigorously analyzed in Ref. [84] and referred to as the low- Q mode.

For excitons in ML TMDCs coupled to dielectric nanocavities, the light-matter interaction strength g can be calculated using the QNMs in combination with material-specific properties of the ML TMDC [59,85]. In this study, we follow Ref. [59] and describe the light-matter interaction in a reaction-coordinate formalism, in which g_{theory} can be expressed as

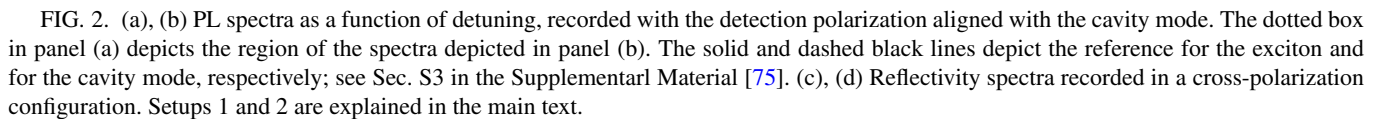
$$g_{\text{theory}} = \sqrt{\frac{\hbar^2 e_0^2}{\pi \epsilon_0 m_0^2 E_{\text{cav}} a_B^2} \sum_{\alpha} \int d^2\mathbf{r} |\tilde{\mathbf{F}}(\mathbf{r}_{xy}, z_{2D}) \cdot \mathbf{p}_{\text{cv}}^{\alpha}|^2}, \quad (1)$$

where $\mathbf{p}_{\text{cv}}^{\alpha}$ denotes the transition dipole moment of the ML TMDC, α denotes the valley index K, K' [65], and a_B denotes the excitonic Bohr radius of the A exciton. The relevant material parameters are extracted from Refs. [33,86–91]. The integral is performed in the *xy* plane of the ML MoTe₂ [cf. Fig. 1(e)], as indicated with the red line in Fig. 1(f). From Eq. (1), it follows that the light-matter interaction

strength strongly depends on the polarization of the cavity mode. If we consider a linearly polarized cavity mode with $\tilde{\mathbf{F}}(\mathbf{r}_{xy}, z_0) = F_0(\mathbf{r}_{xy}, z_0)\hat{e}_y$, then g reaches its maximal value of $\mathbf{p}_{\text{cv}}^{\alpha} \parallel \hat{e}_y$ and vanishes for $\mathbf{p}_{\text{cv}}^{\alpha} \perp \hat{e}_y$. Equation (1) is solved numerically, yielding $g_{\text{theory}} = 5.2 \pm 0.7$ meV; see Sec. S2 in the Supplemental Material [75] for details on the numerical calculations.

We investigate PL spectra as a function of temperature T with the detection polarization $\hat{E}_{\text{det}}$ oriented parallel to $\hat{E}_{\text{cav}}$. Temperature variations primarily affect the excitonic emission energy E_{exc} through the band gap temperature dependence and modify the exciton linewidth Γ_{exc} due to phonon interactions. At $T = 4$ K, we extract $\Gamma_{\text{exc}} = 5.1 \pm 0.9$ meV, in good agreement with Ref. [92]. In contrast, the cavity resonance energy E_{cav} is influenced by the temperature dependence of the refractive index, but its variation is much smaller than the changes in E_{exc} . The cavity linewidth is determined as $\Gamma_{\text{cav}} = 3.3 \pm 0.1$ meV. We explore these dependencies systematically in order to calculate the detuning $\delta = E_{\text{cav}} - E_{\text{exc}}$; see Sec. S3 in the Supplemental Material [75]. for details. Figure 2(a) shows the PL as a function of detuning in a range from $T = 4$ K to $T = 163$ K. In addition, Fig. 2(b) shows the associated line plots in a region of interest close to zero detuning, as indicated in panel (a). In both panels (a) and (b), fits from reference measurements of the uncoupled cavity and of the uncoupled excitons are shown as black dashed and solid lines, respectively. We go from negative detuning to positive values in the measured temperature range, with $T = 40$ K corresponding to $\delta = -0.2 \pm 0.7$ meV. Clearly, avoided crossing is observed, which is a hallmark of strong light-matter coupling.

We use two different setups for different temperature regimes as indicated in Fig. 2(a); see Appendix for details. Setup 1 contains a liquid-helium cryostat for $T \leq 70$ K, corresponding to $\delta \leq 4.9 \pm 0.7$ meV. Setup 2 contains a liquid-nitrogen cryostat for $T \geq 79$ K, corresponding to $\delta \geq 6.2 \pm 1.0$ meV. A small spectral offset is often observed when comparing data acquired with different setups due to imperfect alignment and calibration of the spectrometers. We carefully compare the reference measurements acquired with both setups (cf. Sec. S3 in the Supplemental Material [75]) and deduce an offset of approximately 3 meV between the data from Setups 1 and 2; see Fig. 4 in the Appendix. The data from Setup 2 in Figs. 2(a) and 2(c) have been corrected accordingly. We note that Setup 1 provides a range from $\delta = -3.3 \pm 0.7$ meV to $\delta = 4.9 \pm 0.7$ meV, and therefore, the data from Setup 1 alone already provide evidence of avoided crossing. To assess the light-matter interaction strength, we fit the extracted peak positions of the lower (E_-) and of the upper (E_+) polariton with the real part of a coupled-oscillator model; see Secs. S4 and S5 in the Supplemental Material [75] for details. The fit result, indicated as the red lines in Figs. 2(a) and 2(b), yields $g_{\text{PL}} = 5.3 \pm 0.3$ meV. This value is on the same order as obtained for an ML TMDC on a nanobeam cavity [35,36], despite the much tighter light confinement in this work. This is in line with the prediction that the light-matter interaction strength is largely determined by the out-of-plane confinement [59] and not by the mode volume, as for example, for quantum dots in microcavities [93]. However, as shown in Ref. [62], exciton-exciton interactions are largely determined



The PL measurements are supplemented with reflection measurements in a cross-polarization configuration; see Appendix for details [84]. Figures 2(b) and 2(c) depict reflection spectra as a function of δ . As observed from PL measurements, the avoided crossing is clearly visible as two dips in the reflection spectra, confirming hybridization of excitons and photons. A fit of the reflection measurements with the coupled-oscillator model yields $g_R = 4.7 \pm 0.7$ meV, which agrees well with the value obtained from PL measurements. Notably, the experimentally obtained values g_R and g_{PL} are in excellent agreement with the value obtained with the exciton-reaction coordinate formalism $g_{\text{theory}} = 5.2 \pm 0.7$ meV, as discussed above.

$$N_{\text{Rabi}} = \frac{2E_{\text{Rabi}}}{\Gamma_{\text{exc}} + \Gamma_{\text{cav}}} \geq 1 \Leftrightarrow E_{\text{Rabi}} \geq \frac{\Gamma_{\text{exc}} + \Gamma_{\text{cav}}}{2}, \quad (2)$$

We note that in a coupled-oscillator model, the light-matter interaction strength is in general complex valued [51, 85, 95]. To evaluate the magnitude of the complex part of the

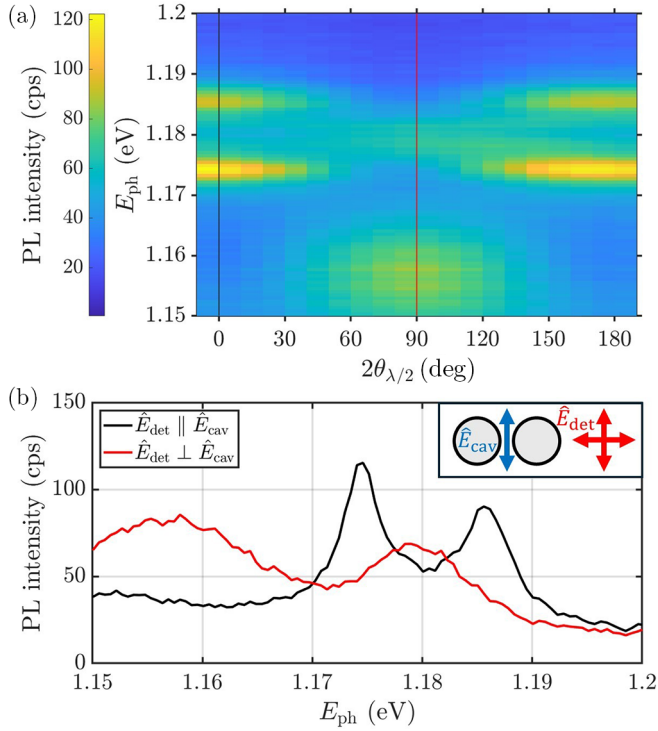


FIG. 3. (a) PL spectra at $T = 50$ K as a function of $\theta_{\lambda/2}$ in front of the analyzer. (b) PL spectra from panel (a) with the analyzer oriented parallel and perpendicular to the cavity mode for black and red lines, respectively.

light-matter interaction strength, we fit the extracted peak positions and the linewidths, corresponding to the real and the imaginary parts of the eigenvalues, respectively; see Sec. S5 in the Supplemental Material [75] for details. We find that the coupled-oscillator model also fits the linewidths accurately, which is additional confirmation of the reported strong light-matter interaction. Moreover, we find that the imaginary part of the light-matter interaction strength is less than 6 % of the real part and is almost zero within the error bars.

Finally, we turn our attention to the polarization properties of the photoluminescence signal. As the cavity mode is linearly polarized, orienting the detection polarization parallel to the cavity mode polarization enables the study of the strongly coupled system. On the other hand, by orienting the detection polarization perpendicular to the cavity mode, primarily

residual excitons that do not couple to the cavity mode are detected. Those are excited in the ML MoTe₂ in the vicinity of the cavity mode. The emission is studied at $T = 50$ K, corresponding to $\delta = 1.4 \pm 0.7$ meV. Figure 3 shows the PL spectra as a function of the angle of the half-wave ($\lambda/2$) plate $\theta_{\lambda/2}$ in front of the analyzer. Together with the analyzer, rotating the $\lambda/2$ plate by $\theta_{\lambda/2}$ effectively rotates the detection polarization state by twice that value. Two peaks, separated by 11 meV and associated with the polaritonic emission, are identified for $\hat{E}_{\text{cav}} \parallel \hat{E}_{\text{det}}$, corresponding to $2\theta_{\lambda/2} = 0^\circ$ and 180° (see Fig. 3). Choosing $2\theta_{\lambda/2} = 90^\circ$ yields a detection polarization perpendicular to the orientation of the cavity mode. One peak centered between the two polaritonic peaks is detected, corresponding to emission from residual excitons that do not couple to the cavity mode. Another peak is observed at $E_{\text{ph}} = 1.157$ eV for $\hat{E}_{\text{det}} \perp \hat{E}_{\text{cav}}$. This is well explained by another mode in the cavity. In Ref. [84], we demonstrate that in addition to the mode of interest in this work, the EDC cavity supports a mode with slightly lower eigenenergy, lower Q factor, and orthogonal polarization. The excitation of this low- Q mode could be due to trions, which are known to exist at a lower transition energy than excitons in the ML MoTe₂ [96].

In summary, we have demonstrated strong coupling between excitons in an ML MoTe₂ and light confined by an EDC cavity on subwavelength length scales. The light-matter interaction strength is deduced both from photoluminescence and reflection measurements, yielding $g_{\text{PL}} = 5.3 \pm 0.3$ meV and $g_{\text{R}} = 4.7 \pm 0.7$ meV. These values are consistent with our numerical calculation exploiting the exciton-reaction coordinate formalism [59], for which we find the value $g_{\text{theory}} = 5.2 \pm 0.7$ meV. Polarization projection of the fluorescence signal confirms the polaritonic nature. This reported system serves as an ideal test bed for studying nanoscale confined light-matter interactions. It combines the small losses of dielectric nanocavities ($\Gamma_{\text{cav}} = 3.3 \pm 0.1$ meV) and subwavelength lateral field confinement ($\sigma \approx 70$ nm $< \lambda/(2n)$). Hence, the polaritons are confined on subwavelength scales by both the out-of-plane confinement of the ML and the in-plane confinement of the dielectric nanocavity. The tight confinement is expected to facilitate strong nonlinearities [57,58,62], and combined with small losses, is expected to give rise to the polariton-blockade effect [60–63]. Moreover, it supports research on condensation effects [50] and chiral interactions [65,67].

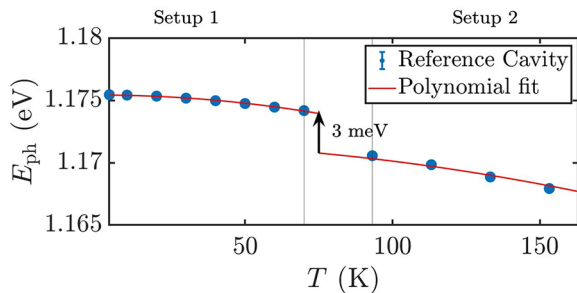


FIG. 4. Extracted peak energy from the reference cavity as a function of temperature. The red lines denote a fit with a parabola. The temperature regions of Setups 1 and 2 are indicated. The deduced offset is marked by the black arrow.

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Data availability. The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

Appendix: Experimental Setups. Two setups are used to access the temperature regime from room temperature to 4 K. Setup 1 contains a closed-cycle liquid-helium cryostat (AttoDRY800XS, Attocube), giving us access to $T \leq 70$ K. Setup 2 contains a liquid-nitrogen cryostat (HFS600, Linkam Scientific), allowing us to measure $T \geq 79$ K. In both setups, the system is excited with a red laser ($\lambda_{\text{laser}} = 650$ and 637.5 nm for Setups 1 and 2, respectively). The excitation light beam is directed via a dichroic mirror (Setup 1) or a 50/50 beam splitter (Setup 2) to a microscope objective, focusing the light on the sample. The emitted light is, after polarization projection with a $\lambda/2$ plate and a linear polarizer (=analyzer), detected by a spectrometer. A long-pass filter ensures the filtering of the excitation light beam. We note that small deviations of the recorded wavelengths are likely to occur when comparing spectra recorded in both setups due to imperfect alignment of the spectrometers. Such an offset is evident in all measurements; see Secs. S3 and S5 of the Supplemental Material [75]. We extract the offset from the reference measurements; cf. Sec. S3 of the Supplemental Material [75]. Figure 4 shows the peak energy of the reference cavity as a function of temperature. The fit of the peak position with a parabola is indicated

as the red lines. A shift in photon energy is observed between $T \leq 70$ K and $T \geq 79$ K, arising from the imperfect alignment of the respective spectrometers used in Setups 1 and 2. The $E_{\text{cav,ref}}(T)$ from Setups 1 and 2 is extrapolated to 75 K, and the offset is calculated as $E_{\text{cav,ref}}^{\text{Setup1}}(75 \text{ K}) - E_{\text{cav,ref}}^{\text{Setup2}}(75 \text{ K})$. This yields an offset of 3 meV (cf. Fig. 4), and the data from Setup 2 in Fig. 2 have been shifted accordingly.

Adaptions to our PL setups allow us to carry out reflection measurements. For that, we change the light source to a superluminescent diode (SLD1050P, Thorlabs). In addition to the analyzer and the $\lambda/2$ plate in the detection path, we insert a linear polarizer and a $\lambda/2$ plate in the excitation path, as well as a quarter-wave ($\lambda/4$) plate in the detection path. In Setup 1, the dichroic mirror is exchanged with a 50/50 beam splitter. The reflection measurements are carried out in a cross-polarization configuration, where the polarization of the incoming light $\hat{E}_{\text{in}}$ is perpendicular to the detection polarization $\hat{E}_{\text{det}}$, for example, $\hat{E}_{\text{in}} = \hat{x}$, $\hat{E}_{\text{det}} = \hat{y}$. The sample is oriented so that the linear polarization of the cavity mode is oriented 45° with respect to both of them, e.g., $\hat{E}_{\text{cav}} = \frac{1}{\sqrt{2}}(\hat{x} + \hat{y})$ [83,84]. The quarter-wave plate is carefully adjusted to suppress elliptically polarized elements induced by scattering at the glass window of the cryostat. All spectra have been normalized with a reference spectrum recorded on a gold surface.

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