Continuous Transition between Weak and Ultra-strong Coupling through Exceptional Points in Carbon Nanotube Micro-cavity Exciton Polaritons

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Non-perturbative coupling of photons and excitons produces hybrid particles, exciton polaritons, which have exhibited a variety of many-body phenomena in various micro-cavity systems. However, the vacuum Rabi splitting (VRS), which defines the strength of photon-exciton coupling, is usually a single constant for a given system. Here, we have developed a unique architecture in which excitons in an aligned single-chirality carbon nanotube film interact with cavity photons in polarisation-dependent manners. The system reveals ultra-strong coupling (VRS up to 329 meV or a coupling-strength-to-transition-energy ratio of 13.3%) for polarisation parallel to the nanotube axis, whereas VRS is absent for perpendicular polarisation. Between these two extremes, VRS is continuously tunable through polarisation rotation with exceptional points separating crossing and anti-crossing. The points between exceptional points form equi-energy arcs onto which the upper and lower polaritons coalesce. The demonstrated on-demand ultra-strong coupling provides ways to explore topological properties of polaritons and quantum technology applications.

Strong light-matter coupling continues to stimulate much interest in diverse disciplines, including quantum optics[1], semiconductor optoelectronics[2], ultrafast optics[3], plasmonics[4], magnetism[5], and circuit quantum electrodynamics[6]. Resonant coupling leads to light-matter hybridisation into two normal modes with an energy separation, $\hbar\Omega_{\mathrm{R}}$, known as the vacuum Rabi splitting (VRS). Different regimes exist, depending on the value of the coupling constant g, with $2g \approx \Omega_{\mathrm{R}}$ when $g \gg \kappa, \gamma$, where κ (γ) is the photon (matter) decay rate. The strong coupling regime is achieved when the cooperativity $4g^2/(\kappa\gamma) > 1$. Furthermore, the ultra-strong coupling regime arises when $g/\omega_0 > 0.1$, where ω_0 is the resonance energy, whereas the deep strong coupling regime is defined as $g/\omega_0 > 1$[7]. In these regimes of light-matter coupling, a variety of phenomena and states are predicted to occur, e.g., a two-mode squeezed vacuum state[8,9], induced by the dynamic Casimir effect[10–12] or through spontaneous conversion[13], unusual photon counting statistics[14], light-matter decoupling[15], and the Dicke superradiant phase transition[16,17].

Micro-cavity polaritons based on Wannier excitons in inorganic semiconductors[18] remain in the strong coupling regime, typically with $g/\omega_0 < 10^{-2}$. Frenkel excitons in organic semiconductors possess large binding energies and oscillator strengths and have displayed larger VRS[19,20]. Moreover, nanomaterials with large binding energy Wannier excitons, such as transition metal dichalcogenides[21] and single-wall carbon nanotubes (SWCNTs)[22,23], have recently emerged, providing a platform for studying strong-coupling physics under extreme quantum confinement. In particular, one-dimensional (1D) excitons in SWCNTs have enormous oscillator strengths, revealing giant VRS in devices containing a film of randomly oriented single-chirality SWCNTs[22].

However, the VRS value in a given exciton polariton system is typically a single constant. One particular class of phenomena that tunable coupling-strength allows one to study is the physics of exceptional points (EPs)[24] in an open or dissipative system with a non-Hermitian Hamiltonian. EPs are spectral singularities where both the eigenenergies and eigenstates coalesce. The rich physics at and around EPs have been demonstrated in diverse physical systems, including microwave cavities[25], coupled atom-cavity systems[26], photonic crystal slabs[27], and exciton-polariton billiards[28]. Exotic phenomena associated with EPs have been observed, including unidirectional transmission and reflection[29], loss-induced revival of lasing[30], and nontrivial geometric phases by encircling EPs[25,28].

Here, we describe a micro-cavity exciton polariton system with built-in coupling-strength tunability and on-demand ultra-strong coupling. The system is based on a cm-sized uniform film of aligned, packed, and single-chirality (6,5) SWCNTs[31,32] in a Fabry-Pérot micro-cavity. The observed value of VRS was up to 329 meV, the highest value ever reported for Wannier excitons, which also corresponds to the largest value ever achieved (13.3%) for the ratio of the coupling strength to the transition energy in a micro-cavity Wannier-exciton polariton system. Interestingly, this large VRS appears only when the polarisation of probe light is parallel to the nanotube alignment direction. For light with perpendicular polarisation, no VRS was observed. This polarisation-dependent VRS allowed us to continuously transition from weak coupling to the strong and ultra-strong coupling regimes through simple polarisation rotation. We observed this behaviour both for the first interband (E_{11}) exciton polaritons in the near-infrared range and the second interband (E_{22}) exciton polaritons in the visible range. Continuous mapping of the polariton dispersion surfaces

revealed four EPs, or two pairs of EPs, symmetrically located with respect to the origin in momentum space, details of which are describable by a model based on quantum Langevin equations. We found two equienergy arcs bounded by each pair of the EPs, and the upper and lower polariton branches coalesce onto the arcs. Furthermore, the VRS exhibited cooperative enhancement, proportional to the square root of the film thickness, quantitative analysis of which determined the E_{11} exciton oscillator strength and dipole moment.

Aligned (6,5) SWCNT micro-cavity-exciton-polariton devices

We utilised the aqueous two phase extraction method[33] to prepare a large-volume high-concentration (6,5)-enriched suspension (Fig. 1a). The suspension was poured into a vacuum filtration system to produce a uniform, optically thick SWCNT film on a polycarbonate filter membrane (Fig. 1b). Through precise control over the filtration process[31], we produced a (6,5)-enriched film that was densely packed and macroscopically aligned (Fig. 1c), with an angle standard deviation of 6.8° in a $\sim$1 cm^2 area (Fig. 1d). Figure 1e shows polarised Raman spectra for the produced aligned (6,5) film, taken with an excitation wavelength of 532 nm, displaying a large difference in the two polarisation configurations. Figure 1f shows absorption spectra, exhibiting strong polarisation dependence. See Methods and Supplementary Information (Supplementary Section 1 and Fig. S1) for more details on the preparation and characterisation of aligned SWCNT films.

Figure 1g shows the basic structure of our aligned SWCNT micro-cavity exciton polariton devices, embedding an aligned (6,5) film with thickness d into a Fabry-Pérot cavity. See Methods

for details on the fabrication procedure. To ensure the strongest possible light-matter interaction, the SWCNT film was positioned at the maximum vacuum electric field position in the cavity. For polariton devices designed to work in the E_{11} region, the SWCNT film was placed in the middle of the cavity to coincide with the field maximum of the fundamental cavity mode (Fig. 1h). For devices optimised for the E_{22} region (Fig. 1i), in order to have a better reflectivity, we used aluminium mirrors instead of gold mirrors. Furthermore, since it was difficult to precisely control the PMMA layer thickness when it was thinner than 100 nm, we utilised the second-order photonic mode of the cavity; the SWCNT film position inside the cavity was close to the field maximum position, as shown in Fig. 1i. We used a Fabry-Pérot cavity with metallic mirrors (as opposed to Bragg mirrors) primarily for simpler fabrication, but its small mode volume was beneficial for the observation of large VRS.

Angle-resolved transmission measurements

By changing the incidence angle (θ in Fig. 2a), we tuned the resonance frequency of the Fabry-Pérot cavity to resonate with the E_{11} or E_{22} of (6,5) SWCNTs. Furthermore, adjustment of the angle between the incident light polarisation direction and the SWCNT alignment direction (ϕ in Fig. 2a) provided a convenient knob to probe the directional variation of the strength of light-matter coupling. The angle θ has a one-to-one correspondence with the in-plane wave vector $k_{\|}$, so the θ dependence of transmission peaks can be converted into k_y or k_x dependence for ϕ = 0° or 90°, respectively, to map out polariton dispersions. Both θ and ϕ were continuously adjustable, as schematically shown in Fig. 2b. The incident light polarisation was kept vertical, which corre-

sponded to s-polarisation with respect to the SWCNT film surface. Rotating the sample around the vertical direction tuned the angle θ, while rotating it around the surface normal varied the angle ϕ. The incident beam size was 2 mm^2, and the transmitted light was collected and analysed with spectrometers equipped with InGaAs and silicon detectors, respectively, for recording transmission spectra in the E_{11} and E_{22} ranges.

Figure 2c shows in-plane dispersion relations along k_y (left) and k_x (right), obtained from transmittance spectra in the E_{11} region for a sample with d = 8 nm at various values of θ for parallel (ϕ = 0°) and perpendicular (ϕ = 90°) polarisations, respectively. All spectra were normalised by the peak transmittance value. A prominent VRS of 135 $\pm$ 9 meV is seen in the dispersion along k_y (left); no splitting is observed in the dispersion along k_x (right). These behaviours are well reproduced by our simulations based on the transfer matrix method; see Fig. 2d. Similarly, in the spectral region of E_{22}, a vacuum Rabi splitting of 170 $\pm$ 10 meV is observed along k_y direction (ϕ = 0°) while no splitting exists along k_x (ϕ = 90°) (see Fig. 2e). Again, the experimental curves are well reproduced by our simulations, as shown in Fig. 2f. See Supplementary Information for a detailed description of the theoretical methods we used, as well as for additional experimental and simulation results (Supplementary Sections 2 and 3, Figs. S2–S10).

Continuously tunable light-matter coupling strength

Furthermore, we can select any value of $\hbar\Omega_{\mathrm{R}}$ between zero and its maximum value, on demand, by setting the polarisation angle ϕ. This is due to the mixing of the parallel and perpendicular

polarisations in the anisotropic SWCNT layer at finite θ; see Supplementary Information (Supplementary Section 4 and Fig. S11) for a detailed theoretical description of the origin of ϕ-dependent VRS. Figure 3a shows a series of transmittance spectra in the E_{11} range for the 8-nm-thick sample at zero detuning ($\theta \sim 39°$) for various values of ϕ from $0°$ (parallel) to $90°$ (perpendicular). Each spectrum was multiplied by a certain value so that the different spectra can be displayed on the same scale. A clear transition from strong coupling (parallel) to weak coupling (perpendicular) is seen. Figure 3b shows the extracted $\hbar\Omega_{\mathrm{R}}$ value for E_{11} as a function of ϕ, quantitatively demonstrating this transition. In the shaded area, the splitting is smaller than the linewidth, and thus, no splitting was observed. The value of VRS for the parallel case, $\hbar\Omega_{\mathrm{R},\parallel}$ = 135 $\pm$ 9 meV, corresponding to $\Omega_{\mathrm{R},\parallel}/(2\omega_{11})$ = 5.5%, where $\hbar\omega_{11}$ is the photon energy of the E_{11} exciton.

The value of $2g$ (and thus Ω_{R}) can be increased by increasing the SWCNT film thickness, d. We demonstrated this by employing a recently developed technique of stacking multiple aligned films[32]. We stacked three layers of aligned (6,5) films, achieving d = 24 nm, and incorporated the stacked film at the centre of the cavity. Figure 3c shows transmittance spectra for this sample at zero detuning for various values of ϕ from 0° to 90°. At ϕ = 0° (parallel), $\hbar\Omega_{\mathrm{R}}$ = 191 $\pm$ 8 meV, corresponding to $\Omega_{\mathrm{R},\parallel}/(2\omega_{11})$ = 7.7%. This value monotonically decreases to 60 $\pm$ 8 meV (or $\Omega_{\mathrm{R},\parallel}/(2\omega_{11})$ = 2.4%) at ϕ = 90°, as shown in Fig. 3d. Our transfer-matrix-based simulation values, also plotted in in Fig. 3d, agree with the experimental values well.

The transmittance spectra taken at continuously adjusted values of ϕ and θ enabled us to map out the full anisotropic dispersion surfaces of the SWCNT exciton polaritons. We used quan-

tum Langevin equations to derive an expression to fit experimental transmittance spectra, with ϕ-independent κ and γ and ϕ-dependent g as adjustable parameters. We then determined these parameters by fitting each spectrum shown in Fig. 3a for different ϕ; the value of $2\hbar g$ using the obtained $g(\phi)$ is plotted against ϕ in Fig. 3b. The values of VRS ($\hbar\Omega_R$) are very close to the values of the coupling strength ($2\hbar g$) in the strong coupling regime, as expected. They start deviating from each other in the transition region from strong coupling to weak coupling. When $\phi > 40°$, $\hbar\Omega_R$ is zero even though $2\hbar g$ remains finite.

Existence of exceptional points in the polariton dispersions

With the obtained values of κ, γ, and g, we calculated the real and imaginary parts of the eigenfrequencies ($\omega_\pm$) using the following equation, which was obtained by solving the characteristic equation of the quantum Langevin equations:

$$\omega_\pm(\theta,\phi) = \frac{1}{2}(\omega_{11} + \omega_{\text{cav}}(\theta)) - \frac{i(\gamma+\kappa)}{4} \pm \sqrt{g(\phi)^2 + \left(i\frac{(\kappa-\gamma)}{4} + \frac{(\omega_{11} - \omega_{\text{cav}}(\theta))}{2}\right)^2}, \quad (1)$$

where $\hbar\omega_{\text{cav}}$ is the photon energy of the cavity resonance. The in-plane wave vector $k_\parallel$ = (k_x,k_y) has one-to-one correspondence with (θ,ϕ). Therefore, we were able to obtain the real part of the eigenfrequencies as a function of (k_x,k_y) for the upper polariton (UP) and the lower polariton (LP), as shown in Fig. 3e for the 8-nm-thick device. See Supplementary Information for a detailed description of quantum Langevin equations, the extraction of eigenfrequencies, and a plot of full dispersion surfaces (Supplementary Section 5 and Fig. S12).

There are four EPs, or two pairs of EPs, symmetrically located with respect to the origin

in momentum space. The points bounded by each pair of the EPs form two equienergy arcs (see Fig. 3f), onto which the upper and lower polariton branches coalesce. Sweeping ϕ leads to a continuous transition between crossing and anticrossing; Fig. 3g shows two dispersions at $\phi = 0^\circ$ (anticrossing) and $\phi = 90^\circ$ (crossing). Furthermore, as shown in Figs. 3h and 3i, we can clearly observe the coalescence of the real part and bifurcation of the imaginary part of the eigenfrequencies as $\hbar g$ is decreased at zero detuning ($\omega_{11} = \omega_{\text{cav}} = \omega_0$), which are hallmarks of EPs in a non-Hermitian system. See Supplementary Information for additional supporting evidence (Supplementary Section 5 and Fig. S13).

Cooperative enhancement of light-matter coupling

We further increased the film thickness to 64 nm to boost the VRS as well as to look for the $\hbar\Omega_{\text{R},\|} \propto \sqrt{N}$ behaviour, where N is the number of dipoles (i.e., excitons in the present case), expected for cooperative light-matter coupling[34,35]. Quantitative analysis of the $\sqrt{N}$ behaviour also allows us to determine the exciton oscillator strength and dipole moment. Figure 4a shows transmittance spectra for the three samples with d = 8 nm, 24 nm, and 64 nm for parallel (ϕ = 0°) polarisation at zero detuning. The VRS increases monotonically with the film thickness. The VRS for the 64-nm sample is $\hbar\Omega_{\text{R},\|}$ = 329 $\pm$ 5 meV, corresponding to $\Omega_{\text{R},\|}/(2\omega_{11})$ = 13.3%, which is the highest value ever reported for micro-cavity exciton polaritons based on Wannier excitons. A similar value has been reported for ultra-strongly coupled Frenkel excitons[36]. The small feature at the high energy end is due to the coupling of cavity photons with the E_{11} excitons of residual (9,1) nanotubes as well as the E_{11} phonon sideband of (6,5) tubes.

Figure 4b summarises the thickness dependence of $\hbar\Omega_{\mathrm{R},\parallel}$ with a linear fit, evidencing the $\sqrt{N}$ scaling expected for cooperative ultra-strong coupling [34,35]. This fit can be compared to

$$\hbar\Omega_{\mathrm{R},\parallel} = 2\hbar\sqrt{\frac{e^2 f_{\mathrm{ex}}}{2n_{\mathrm{c}}^2\epsilon_0 m_{\mathrm{e}} L_{\mathrm{eff}}} \times \frac{N_{\mathrm{C}} d}{\sqrt{3}/2 \times D_{\mathrm{CNT}}^2}} \tag{2}$$

where N_{C} (= 88 nm^{-1}) is the number of carbon atoms per nanometre in a (6,5) nanotube, n_{c} (= 1.483) is the refractive index of the cavity, ϵ_0 is the vacuum permittivity, m_{e} is the electron mass in vacuum, L_{eff} (= 300 nm) is the effective cavity length, and D_{CNT} (= 1.07 nm) is the intertube distance between (6,5) nanotubes. As a result, we obtain the oscillator strength f_{ex} = 0.005 per carbon atom, and the corresponding transition electric dipole moment p_{C} = 0.6 Debye per carbon atom (or 11 Debye per unit cell). See Supplementary Section 6 for more details on the calculations.

Conclusions

These unusually anisotropic SWCNT-based polaritons in the ultra-strong coupling regime can lead to further novel phenomena and devices, including room-temperature polariton condensation and 1D laser diodes. It should be also noted that the vacuum Rabi splitting is comparable to the exciton binding energy, a regime that has only recently been achieved using a traditional GaAs quantum well-based micro-cavity exciton polaritons[37]. The complete and continuous control over the coupling strength addressed, from weak coupling through strong coupling to ultra-strong coupling, can be utilised for investigating fundamental non-Hermitian and topological exciton physics[38,39] and storing and processing quantum information. Finally, the achievement of ultra-strong coupling in the near-infrared and visible range is a significant advantage because sensitive quantum optical methods in the terahertz and mid-infrared are becoming available only recently[40–42].

Methods

(6,5) chirality-enriched aligned nanotube film preparation. Preparation of a (6,5) SWCNT suspension was carried out following prior reports on the aqueous two phase extraction method[33]. The prepared suspension still contained polymers and several different surfactants. In order to have good alignment quality in the final film form, it was crucial to clean and exchange surfactants through ultrafiltration to obtain a monosurfactant aqueous suspension. In addition, the exchanged surfactant concentration had to be kept lower than the critical micelle concentration, which is a necessary condition for spontaneous alignment of SWCNTs to occur during vacuum filtration[31]. Furthermore, thin aligned films were combined to form a thicker film by manually transferring several pieces onto a substrate one by one, while keeping the alignment direction[32].

Device fabrication. A gold (aluminium) mirror was deposited onto a glass slide using a DC sputterer (electron beam evaporator), followed by spinning PMMA on top of it. The whole device was then baked at 170 °C for 5 minutes. The produced aligned (6,5) SWCNT film was first transferred onto a SiO_2/Si substrate by dissolving the polycarbonate filter membrane, following a wet transfer technique[31]. A thin PMMA layer was spin-coated onto the transferred SWCNT film. The spin speed and duration were adjusted to have the second layer PMMA to be of either the same or different thickness compared to the first layer, in order to fabricate polariton devices optimised for the E_{11} or E_{22} region. The SiO_2/Si substrate was then etched away by a potassium hydroxide (KOH) solution, and the PMMA-coated (6,5) film floated on the etchant. The floating film was washed in deionised water several times before it was transferred onto the device, and afterwards, the device was baked again at 170 °C for 5 minutes. Finally, another gold (aluminium) layer of the

same thickness as the first layer was deposited using a DC sputterer (electron beam evaporator).

Data availability statement. 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 W.G. prepared the aligned SWCNT films, fabricated the devices, and performed all measurements under the guidance of J.K. X.L. performed the transfer matrix method simulations. M.B. developed the comprehensive theoretical model to explain the angular dependence of the coupling strength. All authors discussed the results and wrote the manuscript.

Competing Interests The authors declare that they have no competing financial interests.

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Figure 1 Characterisation of macroscopically aligned (6,5) SWCNTs and fabrication of SWCNT micro-cavity-exciton-polariton devices. $\mathbf{a}$, Large-volume high-concentration aqueous suspension of (6,5) nanotubes prepared using the aqueous two phase extraction method. $\mathbf{b}$, Wafer-scale optically thick aligned (6,5) film on a filter membrane prepared by vacuum filtration. $\mathbf{c}$, A scanning electron microscopy (SEM) image of a transferred aligned (6,5) film, showing densely packed alignment structure. $\mathbf{d}$, Angular distribution of (6,5) SWCNTs in an area of $\sim$1 cm^2 with standard angle deviation 6.8°, determined by SEM analysis. $\mathbf{e}$, Raman spectra for a 12-nm-thick aligned (6,5) SWCNT film for two configurations. In the VV (HH) configuration, both the incident and scattered beams are polarised parallel (perpendicular) to the nanotube alignment direction. $\mathbf{f}$, Polarisation-dependent absorption spectra for the same (6,5) film, showing the first and second interband exciton peaks, E_{11} and E_{22}. Several weak absorption peaks are due to the E_{11} and E_{22} of residual (9,1) SWCNTs and the phonon sidebands (PSB) of (6,5) SWCNTs. $\mathbf{g}$, Schematic diagram of the fabricated aligned SWCNT exciton-polariton devices with film thickness *d*. Electric field distribution inside the cavities optimised for the ($\mathbf{h}$) E_{11} and ($\mathbf{i}$) E_{22} region with gold and aluminium mirrors, respectively.

Figure 2 Anisotropic dispersions of micro-cavity exciton polaritons in the E_{11} and E_{22} regions. $\mathbf{a}$, The two angles that are continuously scanned in the present experiments – the incident angle θ and the polarisation angle ϕ. The latter is the angle between the SWCNT alignment direction (*x*-axis) and the incident light polarisation direction. $\mathbf{b}$, Schematic diagram of the experimental angle-resolved transmittance measure-

ment setup. **c**, Anisotropic dispersions of micro-cavity exciton polaritons in the E_{11} region for $\phi = 0°$ and $\phi = 90°$. A clear vacuum Rabi splitting is observed at $\phi = 0°$, while a photon dispersion with no splitting is observed at $\phi = 90°$. **d**, Corresponding simulated anisotropic dispersions in the E_{11} region. **e**, Anisotropic dispersions of micro-cavity exciton polaritons in the E_{22} region for $\phi = 0°$ and $\phi = 90°$, and **f**, corresponding simulated anisotropic dispersions. All transmittance spectra were normalised by the peak transmittance value. Dashed yellow lines indicate the extracted cavity mode (CM) and the E_{11} and E_{22} exciton dispersions.

Figure 3 Dispersions of SWCNT micro-cavity exciton polaritons showing a continuous transition from the weak coupling to strong coupling regime with exceptional points. **a**, Experimental transmittance spectra at zero detuning ($\theta \sim 39°$) for varying polarisation angle ϕ from $0°$ to $90°$ for a device working in the E_{11} region using an aligned (6,5) SWCNT film with d = 8 nm. **b**, The vacuum Rabi splitting ($\hbar\Omega_{\mathrm{R}}$) and the extracted coupling strength ($2\hbar g$) for the device in (**a**) as a function of polarisation angle ϕ. The value of $\hbar\Omega_{\mathrm{R}}$ is continuously tuned from strong to weak coupling. **c**, Experimental transmittance spectra at zero detuning ($\theta \sim 46°$) for varying polarisation angle ϕ from $0°$ to $90°$ for a device working in the E_{11} region using an aligned (6,5) SWCNT film with d = 24 nm. **d**, The vacuum Rabi splitting ($\hbar\Omega_{\mathrm{R}}$) for the device in (**c**) as a function of polarisation angle ϕ. Each transmittance spectrum has been normalised by the respective peak transmittance value. The values of $\hbar\Omega_{\mathrm{R}}$ are larger than those in (**b**) due to the increased film thickness. The simulated values of $\hbar\Omega_{\mathrm{R}}$ show excellent agreement with the experimental

results. Horizontal and vertical error bars in (**b**) and (**d**) are due to the uncertainty of determining polarisation angle ϕ and transmittance peak positions, respectively. **e**, Dispersion surfaces of the upper and lower polartions (colour surfaces) for the device in (**a**). **f**, The equienergy contour at the exciton transition energy. **g**, Dispersions at $\phi = 0^\circ$ and $\phi = 90^\circ$, showing anticrossing and crossing, respectively. **h**, The real and **i**, imaginary parts of eigenfrequencies as a function of $\hbar g$.

Figure 4 Evidence for collective ultra-strong light-matter coupling in aligned (6,5) films and determination of the E_{11} exciton oscillator strength and dipole moment. **a**, Experimental transmittance spectra for $\phi = 0^\circ$ at zero detuning for micro-cavity exciton polariton devices containing aligned SWCNT films of different thicknesses. Each spectrum has been normalised by the respective peak transmittance value. The device containing a 64-nm-thick aligned SWCNT film demonstrates the largest vacuum Rabi splitting 329 $\pm$ 5 meV, the highest value for a Wannier exciton based micro-cavity exciton polaritons. The dips indicated by * and $\hat{}$ are due to the coupling between cavity photons and residual (9,1) SWCNTs, and the coupling between cavity photons and the PSB of (6,5) SWCNTs, respectively. **b**, Vacuum Rabi splitting at $\phi = 0^\circ$ versus the square root of the film thickness, demonstrating the $\sqrt{N}$-fold enhancement of collective light-matter coupling. Vertical error bars are due to the uncertainty of determining transmittance peak positions at zero detuning.