Plasmonic Crystals for Strong Light–Matter Coupling in Carbon Nanotubes

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Supporting Information

ABSTRACT: Their high oscillator strength and large exciton binding energies make single-walled carbon nanotubes (SWCNTs) highly promising materials for the investigation of strong light–matter interactions in the near infrared and at room temperature. To explore their full potential, high-quality cavities—possibly with nanoscale field localization—are required. Here, we demonstrate the room temperature formation of plasmon–exciton polaritons in monochiral (6,5) SWCNTs coupled to the subdiffraction nanocavities of a plasmonic crystal created by a periodic gold nanodisk array. The interaction strength is easily tuned by the number of SWCNTs that collectively couple to the plasmonic crystal. Angle- and polarization resolved reflectivity and photoluminescence measurements combined with the coupled-oscillator model confirm strong coupling (coupling strength ~120 meV). The combination of plasmon–exciton polaritons with the exceptional charge transport properties of SWCNTs should enable practical polariton devices at room temperature and at telecommunication wavelengths.

KEYWORDS: Plasmon–exciton polaritons, plasmonic crystals, single-walled carbon nanotubes, surface lattice resonances, strong coupling

Light–matter interaction is a key aspect of future photonic and optoelectronic devices. Control over the coupling strength between emitters and a cavity allows for the observation of various physical phenomena from spontaneous emission enhancement via the Purcell effect (in the weak coupling regime) to Bose–Einstein condensation. Room-temperature electrically pumped polariton lasing, an all-optical polariton transistor, and nonclassical light generation in the strong coupling regime. The relation between the average dissipation rate in the system (γ) and the exciton–cavity energy exchange rate—in the Rabi splitting (ℏΩ)—leads to a transition from the weak (γ ≪ ℏΩ) to the strong (γ ≳ ℏΩ) coupling regime. The Rabi splitting can be controlled by the number of emitters (N) that collectively couple to the cavity, their oscillator strength (f), and the cavity mode volume (V_m) as ℏΩ ∝ √N/V_m.

For typical Fabry–Pérot cavities the large mode volumes are challenging for applications requiring miniaturization and/or operation on the single emitter level in low-energy optoelectronic devices. As an alternative to that, 2D plasmonic crystals supporting hybrid photonic–plasmonic nanocavities have been recently used to investigate and exploit strong light–matter coupling. Plasmonic crystals represent a class of structures formed by periodically patterned metallic structures, e.g., nanoparticle or nanohole arrays, and combine the advantages of high quality factors of conventional photonic cavities and the nanoscale field localization of plasmons. The characteristic period of these structures is comparable to the wavelength of the plasmon resonances (e.g., local plasmon resonances, LPRs) that allows diffractive or waveguide coupling to create surface lattice resonances (SLRs) or quasi-guided modes, respectively. In order to tailor the dissipation rate and field confinement, the photon–plasmon composition of the final modes is varied via the periodicity and size or shape of the metallic nanostructures.

With regard to suitable materials, strong light–matter interaction at room temperature requires a large exciton binding energy and hence is limited to large bandgap inorganic semiconductors, e.g., GaN5,16 and ZnO,17 monolayered MoS2,19 or MoSe2,20 or organic materials with their strongly bound Frenkel excitons. Recently, we reported that semiconducting single-walled carbon nanotubes (SWCNTs)20 in microcavities also show strong light–matter coupling and thus formation of exciton–polaritons at room temperature. Their unique optoelectronic properties offer a number of key advantages. First, SWCNTs exhibit a small Stokes shift and narrow excitonic transitions (~20–40 meV) which is a result of their large exciton oscillator strength. Second, the high exciton binding energy (~300 meV) and exceptional charge transport properties of SWCNTs as well as their chirality-dependent excitonic transition energies offer the opportunity for broadly tunable electrically pumped polariton devices at...
room temperature. Finally, as narrow-bandgap semiconductors (first excitonic transition: $E_{\text{ex}} \sim 0.5\text{−}1.3$ eV) SWCNTs are also attractive candidates for emission in the telecommunication wavelength range and for investigation of the ultrastrong coupling regime, for which Rabi splitting is comparable to the uncoupled transition energies.\textsuperscript{27−30} The full potential of ultrastrong coupling is still unexplored, but it is considered to lead to new intriguing phenomena such as quantum vacuum radiation\textsuperscript{31} and quantum phase transitions.\textsuperscript{32} The integration of SWCNTs with plasmonic crystals may also present an important step toward the long-standing goal of achieving an electrically pumped carbon-based (plasmon)−exciton polariton laser. It was recently reported that an admixture of the plasmonic component in compound plasmon-exciton polaritons (PEPs) reduces their effective mass and facilitates room temperature quantum condensation because the critical temperature for condensation is inversely proportional to the effective mass.\textsuperscript{12,33}

Previously, we demonstrated that in the weak coupling regime semiconducting (6,5) SWCNTs coupled to plasmonic crystals can be turned into broadband tunable emitters by exploiting the Purcell effect. Here, we explore the evolution of plasmon-exciton interactions in the strong coupling regime by increasing the number of SWCNTs coupled to square lattice plasmonic crystals. Using the coupled-oscillator model we describe the experimentally observed angle-dependent reflectivity and photoluminescence (PL) data. We find a coupling strength of up to $\sim 120$ meV, which corresponds to light−matter interaction approaching the ultrastrong coupling regime.

Figure 1c shows the absorbance spectra of three SWCNTs films with different thicknesses. The spectra are dominated by the $E_{22}$ and $E_{11}$ transitions of (6,5) SWCNTs at $\lambda = 575$ nm and $\lambda = 1000$ nm, respectively, confirming the presence of mainly a single SWCNT species. The line width of the $E_{11}$
transition is only 48 meV. The $E_{11}$ absorbance increases superlinearly with the thickness of the SWCNT layer (see Supporting Information Figure S1), indicating a densification of the SWCNT layer during the successive drop-casting process, which is favorable for improving coupling to the nanocavities. The emission of our SWCNT layers is also dominated by the exciton transitions of (6,5) SWCNTs ($\gamma_{ex} = 47$ meV; see photoluminescence-excitation map in Supporting Information Figure S2).

The scanning electron micrograph of the electron-beam lithographically fabricated NDs (area $100 \times 100 \mu m^2$) in Figure 1d illustrates the square symmetry of the array and shape of NDs (slightly elliptic, with diameter $160-180$ nm). The square lattice was selected as it represents one of the simplest cases of a 2D periodic structure facilitating interpretation of the band structure of the plasmon–exciton polaritons. The corresponding first Brillouin zone is shown in the inset of Figure 1d, where the three high-symmetry points ($\Gamma$, $X$, $M$) are indicated. The pitch of the ND array was chosen such that the spectral position of the $\Gamma$-point was at approximately $\lambda = 1005$ nm ($\lambda \approx n \times \sin(\theta)$) and thus at the minimum detuning with the $E_{11}$ transitions of the (6,5) SWCNTs at 1000–1010 nm. The theoretical dispersion relations of the plasmonic crystal along the $\Gamma$–$M$ and $\Gamma$–$X$ directions are shown in Figure 1e. The polar angle scale (i.e., $\sin(\theta)$) is used for direct correlation with the experimental angle-dependent optical response. The dispersion shows that the nanocavity modes are related to the $(\pm 1,0)$ and orthogonal $(0,\pm 1)$ Rayleigh anomalies (RAs). The corresponding relation is given by

$$\frac{2\pi n}{\lambda} = \sqrt{(k_0 \sin(\theta) \cos(\varphi) + i 2\pi/a)^2 + (k_0 \sin(\theta) \sin(\varphi) + j \times 2\pi/a)^2}$$

(1)

where $k_0$ is the free space wavevector, $n$ the refractive index in the region surrounding the ND array, and $i$, $j$ are the RAs orders, e.g., 0, ±1, ±2, and so forth. The azimuthal angles for the $\Gamma$–$M$ and $\Gamma$–$X$ directions are $\varphi = 0$ and $\varphi = \pi/4$, respectively. As one can see, some modes are degenerate, e.g. $(0, \pm 1)$ for the $\Gamma$–$X$ direction and the pairs $(−1, 0)$, $(0, −1)$ and $(+1, 0)$, $(0, +1)$ for the $\Gamma$–$M$ direction.

Upon coupling of the LPRs supported by the gold NDs to these diffraction orders the hybrid photon–plasmonic SLRs appear. The corresponding energy exchange leads to the renormalization of the final nanocavity modes. In order to investigate the band structure of the SLRs, we performed angle-resolved reflectance measurements of a sample without SWCNTs for $\varphi = 0$ and $\varphi = \pi/4$ (Figure 1f). We used a Fourier imaging configuration where the orientation of the entrance slit of the spectrometer defined the plane of detection.
and thus the azimuthal angle (see Supporting Information for details). The dispersion extracted from the reflectivity data (i.e., maximum intensity for each angle of detection) was fitted to a coupled-oscillator model (white lines), taking into account the dispersion of the RAs and spectral position of the LPRs along the shorter or longer axis of the NDs (see Methods and Supporting Information, Figure S3). For the \( \Phi \)-point and thus the in-plane wavevector component \( k_{\parallel} = 0 \) both curves split and form a photonic stop band that indicates coupling between two RAs and lifting of the degeneracy.\textsuperscript{37,38} We note that, for the coupled-oscillator model used to fit SLRs dispersion, we only used data for the lower SLR branch (L-SLRs). No reliable experimental data can be extracted for the upper SLR branch (U-SLRs) due to the Fano-like interaction between broad LPRs and narrow RAs, which results in extremely weak reflectivity for the U-SLRs. This also led to the slight discrepancies (\( \sim 10 \text{ nm} \) at \( \Gamma \)-point) in the fitted curves for the \( \Gamma-M \) and \( \Gamma-X \) directions.

Upon introducing the SWCNTs to these nanocavities the excitons effectively couple to the upper and lower SLRs (U-/L-SLRs) and form upper, middle, and lower plasmon–exciton polaritons (U-/M-/L-PEPs) as illustrated in Figure 2a. To demonstrate this experimentally, we performed angle-resolved reflectivity and PL measurements on ND arrays coated with SWCNT layers of different thicknesses (see Supporting Information for details). Additionally, the transverse-electric (TE) or transverse-magnetic (TM) light polarization were selected to discriminate the spectral features as depicted in Figure 2b.

Figure 2c and d presents the angle- and polarization-resolved PL for samples with SWCNT layers of different thicknesses (\( h = 100, 200, \) and 300 nm) for \( \Phi = 0 \) and \( \Phi = \pi/4 \), respectively (see Supporting Information Figure S4 for the reflectivity data). The common dispersionless spectral feature at \( \Lambda = 1000-1020 \text{ nm} \) corresponds to the excitonic emission of the SWCNTs with an increased Stokes shift for thicker layers. This signal corresponds to the share of SWCNTs not coupled to the nanocavities. In order to estimate the coupling strength between excitons and SLRs (white dotted lines in Figure 2c and d), we extracted PL maxima and fitted those to the eigenvalues of the effective Hamiltonian:\textsuperscript{12}

\[
H = \begin{pmatrix}
E_{ex} & V_{U-SLR} & V_{L-SLR} \\
V_{U-SLR} & E_{U-SLR}(\theta) & V_{U-L} \\
V_{L-SLR} & V_{U-L} & E_{L-SLR}(\theta)
\end{pmatrix}
\]

where the diagonal terms \( E_{ex}, E_{U-SLR}(\theta), \) and \( E_{L-SLR}(\theta) \) correspond to the uncoupled exciton, the upper, and the lower SLRs states, respectively. The off-diagonal terms \( V_{U-SLR} \) and \( V_{L-SLR} \) represent exciton-SLR coupling energies and are used as the fitting parameters. In order to allow the fit to converge, we also included an additional coupling term \( V_{SLR} \) reflecting interaction between U-SLR and L-SLR. Although its value increases for thicker SWCNT films (up to \( \sim 80 \text{ meV} \)) and thus seems to be affected by the excitons, the mechanism of this coupling is not clear yet as the direct exciton-SLR coupling is already included as \( V_{U-SLR} \) and \( V_{L-SLR} \). The resulting dispersion curves for the M-PEP and L-PEP show a reasonably good fit (Figure 2c,d, left and middle, white solid lines). For the sample with the thinnest SWCNT layer (Figure 2c,d, left) only the L-PEP branch can be resolved due to the weak far-field response of the U-SLRs as discussed above. The same applies to the reflectivity data (see Supporting Information, Figure S4). However, for the thicker SWCNT layers (Figure 2c,d, middle and right), the M-PEP, which originates from splitting of the U-SLR, is also visible, while the U-PEP remains undetectable. In addition, the TE-polarized M-PEPs at \( \Phi = \pi/4 \) show an apparent bending behavior for \( \sin(\theta) > 0.4 \) (the data are not taken into account during the fitting procedure, since they originate from the \((-1, 1)\) RAs that are clearly visible in this range of the uncoupled state (indicated with red arrows in Figure 2d). It is also important to note that the U-SLR for TE polarization at \( \Phi = 0 \) and TE/TM at \( \Phi = \pi/4 \) as well as the associated U-PEP and M-PEP at \( \sin(\theta) = 0 \) are optically inactive due to the symmetry of the field distribution around the NDs.\textsuperscript{35,39} However, they can be changed from dark to bright by varying the ND shape and pitch of the array.\textsuperscript{10} The same effect also prevents an observation of these states in TM polarization for all angles of detection. This behavior has an important implication for lasing because it corresponds to the reduction of radiative losses as highlighted recently.\textsuperscript{13}

We note that our model does not include nontrivial spectral features (indicated with white arrows in Figure 2c, middle and right) for the TE-polarized M-PEP at large angles (that is \( \sin(\theta) > 0.3 \)), and thus these data points were excluded from the fit. These features are more pronounced in a sample with the larger exciton-SLRs detuning, i.e., ND array pitch \( a = 830 \text{ nm} \) (see Supporting Information, Figure S5). As for their origin we have to take into account that this branch-like feature is optically inactive at small angles as is the TE-polarized M-PEP. A closer look also reveals that it is convex (i.e., \( d^2I/d\Omega k < 0 \)) which resembles the TM-polarized M-PEP. It seems that
although the TE-polarized dipoles should not be able to couple to the orthogonal RAs this process is possible due to the one-dimensional structure and strongly anisotropic polarizability of the SWCNTs. In other words, for the random network of SWCNTs the off-diagonal terms in the polarizability tensor are nonzero and thus cross-coupling between TE- and TM-polarized LPs and RAs is allowed. The discussed SWCNTs-mediated TE-TM cross-coupling mechanism is probably also responsible for the lifted degeneracy of the L-PEPs for $\varphi = \pi/4$ (Figure 2d, middle and right). For samples with higher SWCNT concentrations the L-PEP branch splits into TE and TM-polarized emission where the slight secondary appearances indicated with white arrows are visible. To quantify and visualize the changes of band structure of the PEPs for different amounts of SWCNTs more clearly, Figure 3a and b summarize the dispersion curves and the experimental data used for the fit at $\varphi = 0$ and $\varphi = \pi/4$, respectively. The energy separation between U-PEP and M-PEP for the angle at which $E_{\text{ex}}$ equals $E_{\text{USLR}}$ progressively increases with increasing thickness of the SWCNT film from around 25 to 300 meV. In general, a good correlation is found between the calculated PEP dispersions and the experimental data despite the simple coupled-oscillator model used here. However, there are some deviations of the TE-polarized emission of the L-PEP for the sample with the thickest nanotube layer (Figure 2c,d right). The main fitting simplification causing these imperfections is the constant mode profile (and thus coupling strength) for different angles over a large spectral range as well as its overlap with the optically anisotropic SWCNTs layer. Moreover, the discussed SWCNT-mediated interaction between TE-TM-polarized PEPs was not included in the model.

The dependence of the coupling strength on the number of SWCNTs in the layer helps to justify the collective effects and thus light–matter interaction. Figure 4 shows a summary of the exciton-USLR coupling term in eq 2 for $\varphi = 0$ and $\varphi = \pi/4$ (open and solid symbols) and for TE (black) and TM (red) polarization (see Supporting Information, Figure S7 for a summary of all coupling terms, i.e., $V_{\text{USLR}}$, $V_{\text{LSLR}}$, and $V_{\text{UL}}$). The data are presented as a function of the square root of the effective absorbance $= \sqrt{\frac{\Delta \nu_{\text{II}}}{2}} (\langle EF \rangle - 1)$, where the halved absorbance $\left(\frac{\Delta \nu_{\text{II}}}{2}\right)$ is proportional to the average number of SWCNTs oriented in one direction and the field intensity enhancement factor $\langle EF \rangle$ is the average over the whole volume of SWCNTs around the gold NDs (see Supporting Information, Figure S6). The latter characterizes the spatial overlap between the SWCNTs and the mode volume of SLRs, or in other words the efficiency with which SWCNTs couple to the SLRs. An enhancement factor of 1 (i.e., no enhancement) is subtracted to exclude the unphysical increase in coupling for a thicker layer. The observed trends show a clear dependence on the thickness of the SWCNTs layer as expected for the strong coupling regime. The error bars in Figure 4 reflect the uncertainty and variation of the layer thickness. The additional horizontal dashed lines indicate the boundaries between the three coupling regimes, i.e., weak, strong, and ultrastrong. The transition from weak to strong coupling can be approximately defined via the coupling strength ($V$) as $V > \sqrt{\gamma_{\text{ex}}^2 + \gamma_{\text{SLR}}^2}/8$, where $\gamma_{\text{ex}}$ and $\gamma_{\text{SLR}}$ are the line widths of the excitonic transitions ($\gamma_{\text{ex}} = 47$ meV) and SLRs ($\gamma_{\text{SLR}} = 55$ meV), respectively. For the sample with the smallest amount of SWCNTs, the estimated values are in the transition region, while strong coupling is clearly present for the sample with the thicker layer. The larger spread of values for the thinnest layer is explained by the lack of the experimental data for the M-PEPs. With respect to this, it may be also more reliable to illustrate the transition between regimes via the coupling term between exciton and L-SLRs. Although this nanocavity mode is slightly detuned compared to the U-SLRs, the coupling strength still indicates a strong coupling regime (see Supporting Information, Figure S7). Increasing the amount of SWCNTs around the gold NDs further, leads to another transition, from strong to ultrastrong coupling ($V > 0.1E_{\text{ex}}$). Although the ultrastrong coupling regime was not reached completely and no new effects were observed, further increase of the coupling strength may reveal interesting phenomena. This could be achieved, for example, via the incorporation of SWCNTs with a larger diameter and thus smaller transition energies or by aligning the carbon nanotubes in one direction and thus increasing their effective density. The need of a large number of SWCNTs for the observation of collective strong light–matter coupling explains the lack of corresponding effects in previous reports dealing with single SWCNTs or relatively thin films coupled to microcavities.

In summary, we have shown the progressive increase from the weak to the strong and up to the ultrastrong coupling regime between (6,5) SWCNTs and plasmonic polaritons supported by a periodic gold nanodisk array. Aside from exploring SWCNTs as a promising material for strong light–matter coupling at room temperature, our hybrid plasmon–exciton polariton structures could be easily integrated in optoelectronic devices (e.g., light-emitting field-effect transis-

![Figure 4](https://example.com/figure4.png)

**Figure 4.** Coupling strength between excitons and upper surface lattice resonance as a function of the square root of the effective absorbance (i.e., taking into account the $E_{\text{ex}}$ absorbance and volume-averaged coupling efficiency) for TE (black) and TM (red) polarized light and detection at $\varphi = 0$ (open symbols) and $\varphi = \pi/4$ (solid). Transition regions of weak-to-strong and strong-to-ultrastrong coupling regime are indicated with horizontal, dashed lines. Least squares fit to the values is presented with dotted lines. For representation of all coupling strengths see Supporting Information S7.
take advantage of SWCNTs with their large oscillator strength and high charge carrier mobilities.

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**Author Contributions**
The manuscript was written through contributions of all authors. Y.Z designed the experiments, carried out sample characterization, and performed theoretical analysis. A.G. prepared monochiral SWCNTs dispersions, designed, and built the angle-resolved optical setup and helped with measurements. J.Z conceived and supervised the project. All authors have given approval to the final version of the manuscript.

**Notes**
The authors declare no competing financial interest.

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