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Carbon Nanotubes

Wiry matter-light coupling

Electrical-injection has been achieved into polaritons built from admixing excitons in carbon nanotubes with light in a surrounding microcavity.

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Coaxing light and matter to interact strongly enough to fully mix into coupled states has been the focus for scientists bridging photonics, materials, and chemistry over the past three decades. Admixing states creates new quasiparticles with unusual material properties. Their pursuit is relevant to efficient low-threshold lasers, nonlinear optical materials for switching that outperform anything previously available, and solid-state Bose-Einstein condensates that underpin quantum technologies. Writing in Nature Materials, Jana Zaumseil and colleagues have developed a way to incorporate size-selected carbon nanotubes into micron-sized photonic cavities, fully mixing the excitons with light at room temperature but also, crucially, demonstrating their simple electrical excitation.[1]

To access such 'strong-coupling' an emitter (typically a semiconductor exciton) is confined inside an optical cavity so that both the confined photon and exciton are in resonance. When exciton emission and absorption are fast enough, the energy oscillates back and forth creating two split energy states called polaritons which are part-light, part-matter, with unusual new properties emerging. First observed at low temperatures in planar microcavities of GaAs-based quantum wells, many material systems have now shown polaritons at room temperature including wide-bandgap semiconductors (GaN, ZnSe, ZnO),[2] organic semiconducting polymers and small dye molecules which allow simple low-cost processing[3,4], and more recently transition-metal dichalcogenides (TMDs).[5] Even single quantum dots or individual molecules[6] can yield strong coupling, opening prospects for manipulating photons one at a time.

One promise of polaritonics is achievement of low-threshold lasing, which does not rely on inversion of the excited state population but on the bosonic nature of the lower polariton state. Instead of needing to scale down the device size (so that inverting all emitters costs less energy), this offers a different paradigm for coherent light emission, using the stimulated scattering of polaritons into their lowest state where they condense into a macroscopic condensate. While a number of polariton lasers have worked with optical pumping, this is unsatisfactory for real devices and a key goal has been incorporating electrical injection into such microcavities.

The combination of high-oscillator strength excitons in small optical cavities creates problems for wiring up such devices (Figure 1). If the electrons overlap the cavity photons, the extra free-carrier absorption often ruins the strong coupling, so that conventional LED structures have to incorporate trade-offs that yield polaritons but no condensation (Figure 1a).[7] On the other hand sideways injection of the electrons has required complex fabrication and highly compromised coupling (Figure 1b).[8,9] Even electrical control of the polaritons has been challenging, though now advanced to the stage where ultra-low-energy switching is possible at low temperatures.[10] This is where Zaumseil and coauthors show a significant advance, using a carbon-based material system not previously considered for polaritonics.

Single-walled carbon nanotubes support tightly-bound strongly light-emitting excitons at room temperature and around telecommunications wavelengths, but because their energies depend on how the nanotubes roll up, the required narrow emission lines can only be obtained by size-selection. Laying down oriented mats of these nanotubes and electrically contacting from each side allows both electrons and holes to be injected from opposite ends (Figure 1d). When these recombine in the central zone of a nanotube, they form excitons which emit light. A simple planar cavity formed of mirrors above and below the nanotubes multiply reflects the light, giving exciton-photon strong coupling. Very elegantly, the one metallic mirror can be used as a voltage gate that shifts the lateral position of the polariton emission within the microcavity, as well as controlling the polariton energies.

While strong polariton emission is seen in these devices, condensation is not yet observed at high injection currents. A key aspect is the formation of the runaway population of the lower polariton by stimulated scattering from the injected reservoir of hot excitons. Both optical phonons and Coulomb-induced exciton-exciton scatterings contribute, and enhancing the latter is critical. In carbon nanotubes (and other systems with high exciton binding energy), the Bohr radius of excitons is on the nanoscale with centre of mass delocalisation below 100nm which reduces the exciton overlap and thus the Coulombic in-scattering. This suggests that high densities will be required for lasing. Applying vertical fields can polarise the excitons, producing 'dipolaritons' (superposition of a photon, a direct and an indirect exciton) with much larger Coulombic scattering,[11] of great interest in stacked TMD systems, and also potentially of interest in low dimensional systems such as nanotubes. Another promising approach just emerging is the use of plasmonic resonators, which confine light millions of times more tightly than conventional microcavities while retaining polariton splittings exceeding thermal energies.[6] This can strongly enhance the Coulombic scattering and potentially open the way to realistic nonlinear optical devices for optical and quantum processing.

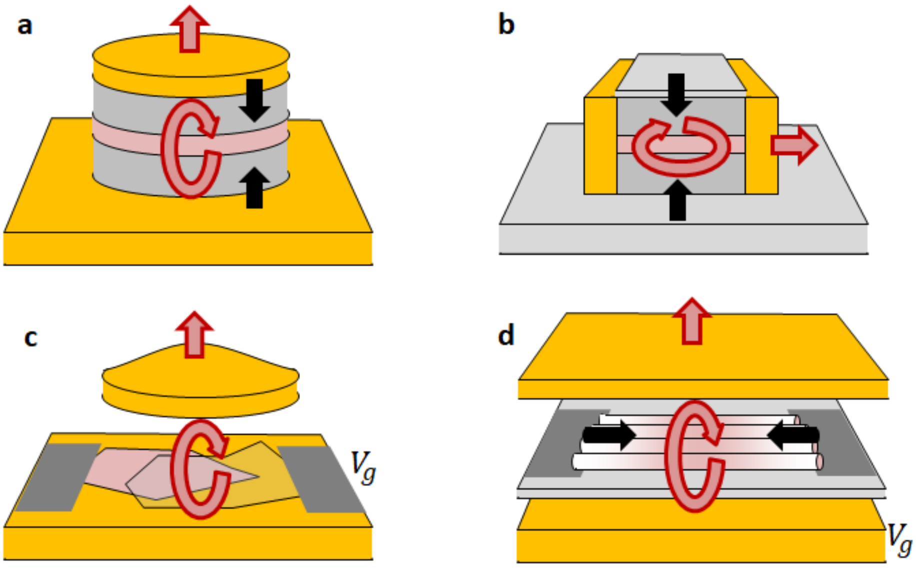

Figure 1 | Strategies for electrical injection into strong-coupling microcavities. a, Conventional planar semiconductor microcavity, patterned into a mesa with top and bottom electrical contacts (black arrows) through the mirrors (yellow) which also confine the light vertically (red arrows). **b**, Side-clad mirrors around vertical LED gain structure. **c**, Monolayer TMD semiconductor microcavity using graphene gate (V_g) inside top micromirror and bottom mirror. **d**, New scheme using carbon nanotubes which are side-contacted to generate light emission from the cavity centre, with light emission vertical.

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