STRONG EXCITON-PHOTON COUPLING IN LOW-Q MICROCAVITY

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Abstract: Here we demonstrate the optical features of fabricated inorganic-organic hybrid multiple quantum well (IO-MQW) structure, Cyclohexenyl ethyl ammonium lead iodide, (C₆H₄C₄H₄NH₃)₂PbI₅ (CHPI). These hybrids possess strongly-resonant optical features, thermally stable and are compatible with hybrid photonics assembly. Room-temperature strong-coupling is observed when these hybrids are straightforwardly embedded in metal-air (M-A) and metal-metal (M-M) low-Q microcavities, due to the large oscillator strength of these IO-MQWs. The strength of the Rabi splitting is 130meV for M-A and 160meV for M-M cavities.

1. INTRODUCTION

The perspective of achieving a better control over the spontaneous emission in optoelectronic devices has been a major momentum to research in photonic structures including microcavities. Confining emitters within microcavities modifies their emission in two related regimes; weak coupling and strong coupling. In weak coupling, the spatial and the temporal distribution of the emitted radiation is modified and hence the spontaneous emission is enhanced or suppressed. In contrast for larger light-matter coupling, if exciton and photon lifetimes are long in comparison to their interaction time, the interaction comes in strong coupling regime in which new coupled eigenstates called polaritons emerge. In this regime, the microcavity polariton dispersion $E(k)$ consists of anti-crossing branches separated in energy by the Rabi splitting, $\Omega$, which defines the coupling strength. Although exciton-photon coupling in such strong-coupling cavities is of great interest, most studies focus on using inorganic emitters (semiconductor Quantum Dots (QDs) and Quantum Wells) in high Q-factor planar microcavities at low temperatures. Organic semiconductors are most promising alternatives as laser media and indeed strong Frenkel exciton-photon coupling has now been realized even at room temperatures. Recently we reported strong coupling in low Q- microcavities containing organic layers (of J-aggregates). However, trivial problems related to organic semiconductors, such as low-melting values, device-compatible processing issues, and photo-oxidation are limiting factors for practical applications [1-4].

Recent progress in the structural engineering of ‘naturally’ self-assembled 2D layered inorganic-organic multiple quantum wells (IO-MQW) show new possibilities for optoelectronics devices with tremendous advantage over the conventional organics. These metal-halide-based IO-MQWs are thermally stable up to 200°C, and a wide range of structures can be conveniently synthesized by choosing an appropriate intercalating organic moiety. While fabrication of well-ordered thin films is not always straightforward, we recently reported a simple intercalation strategy, compatible with fabrication of photonic devices, to yield well-ordered films over cm²-scale areas [5-7].

Here we fabricate large area heterostructures from a self-assembled hybrid quantum well, 2-(1-Cyclohexenyl) ethyl ammonium lead iodide, (C₆H₄C₄H₄NH₃)₂PbI₅ (hereafter CHPI). We show the ability of these hybrids to generate the strong coupling regime even at room temperatures in low-Q factor microcavities.

2. EXPERIMENT

Thin films and single crystals of CHPI were prepared by both intercalation and chemical synthesis described previously [7, 8]. The resulting orange precipitate was filtered, dried and used to harvest single crystals of dimensions 2x2x0.5mm³ from methanolic solution by slow evaporation. X-Ray diffraction confirms the natural self assembly of layered films. Microcavities were fabricated as follows: firstly silver is sputtered onto a clean glass substrate, followed by sputtering of a SiO₂ layer on top of the Ag. Subsequently PbI₂ was coated onto this structure and subsequently processed to obtain CHPI through intercalation, as described earlier. This forms a metal-insulator-CHPI-air (M-A) microcavity. A layer of poly(methyl methacrylate) (PMMA) polymer was spun on top of the CHPI and subsequently a partially-reflecting Ag mirror of thickness was coated by thermal vapor deposition to form metal-insulator-CHPI-insulator-metal (M-M) cavity. PL and transmission imaging/spectroscopy of were carried out using a modified laser scanning confocal microscope fitted with a diode laser (447nm), white light source and a spectrometer.

3. RESULTS AND DISCUSSIONS

ICOP 2009-International Conference on Optics and Photonics
CSIO,Chandigarh,India,30 Oct.-1 Nov.2009
In these hybrid 2D layered networks, inorganic semiconductor and organic layers are stacked alternatively. These are regarded as natural multiple quantum well (MQW) structures, with ~6Å semiconductor layers as ‘well’ material and wider-bandgap ~10Å organic layers as the ‘barrier’. Strong room-temperature excitons in these heterostructures arise due to the low dimensionality of the inorganic structure (quantum confinement), as well as large dielectric mismatch (dielectric confinement) between the organic and inorganic layers. Films processed using the intercalation strategy show narrow X-ray diffraction peaks [Fig. 1(A)] corresponding to the (001) plane at 2θ = 5°, followed by higher diffraction orders related to (00l) (l=2, 3, 4, ...) thus confirming that the CHPI films are highly self-organized into layers and perfectly orientated on the substrate. These films show room-temperature excitonic absorption at ~510nm and strong photoluminescence (PL) at ~523nm [Fig. 1(B)]. Both absorption and PL show symmetric narrow lineshapes with FWHM less than 20nm.

It is essential to examine the nature and morphology of the CHPI thin films prior to their integration into microcavities, since the PL characteristics are strongly dependent on structural re-arrangement of layers. Figure 2(A) gives the morphological study through PL intensity mapping of a micron sized single crystal. As the thickness of the crystal is in the order of microns the transmission image is totally saturated at the peak exciton absorption wavelength 510nm (not shown here). PL mapping of the single crystal shows strong and narrow exciton PL peak at 523nm, uniformly throughout the crystal. However, a broad shoulder at about 545nm has also been observed at the crystal edges. The uneven packing of layer stacking at the edges of the crystal may result into more crystal defects, thus the defect related broad emission at 545nm.

In contrast to measurements on the single crystals, in thin films the PL mapping is spatially uniform and no such defect-related emission has been observed. However, there is a PL red-shift (~10nm) [Fig. 2(B)] when the thickness is increased from 40nm to 350nm. This shift may arise probably due to distorted bond angles within the lead iodide layered network. From the PL mapping measurements, it was concluded that ~100nm thick films of CHPI are optimal to produce the strong coupling regime.

Having optimized the fabrication process for uniform thin CHPI films by intercalation strategy they are conveniently placed within low-Q microcavities. The first M-A cavity is designed as a 5/4 thickness cavity, with Ag/SiO2 (R=86%) and CHPI/air as high and low reflecting ends. The SiO2 spacer thickness is adjusted so that the cavity mode is in resonance with the exciton state. Angle-resolved transmission spectra of the M-A cavity are recorded using TE-polarized white light. The photonic mode at 2.3eV (540nm) of width ~0.12eV (~70nm) splits into two branches at the exciton transmission dip around 2.45eV (523nm), at an incident angle of 33° [Fig. 3(A)]. To confirm this, full-transfer-matrix simulations, are performed using the experimentally-extracted optical constants (n and k) of CHPI from white-light ellipsometry, and the values reported in literature for Ag and SiO2 over the wide region of wavelengths, λ=300-800nm. Both simulated and experimental transmission spectra clearly reveal characteristic anti-crossing transmission dips from strong-coupling of the excitonic and cavity modes forming two new branches, lower polariton (LP) and upper polariton (UP) branches [Fig. 3(B)]. The vacuum Rabi splitting defined by the minimum energy separation between the LPB and UPB and is calculated to be ~130meV. This anti-crossing is a clean signature of strong coupling between CHPI hybrid exciton and the cavity photonic mode.

The dispersion relations of the polaritonic branches also match the standard two-level model with UPB (E_u) and LPB (E_l) defined by the following equation

\[ E_{u,l}(\theta) = \frac{E_{ph}(\theta) + E_{CHPI}}{2} \pm \sqrt{\frac{\Omega^2}{4} + \frac{E_{ph}(\theta) - E_{CHPI}}{4}} \]
where $E_{\text{CHPI}}$ and $E_{\text{ph}}$ are the energies of CHPI excitonic state and the bare cavity photon mode respectively. The cavity photon mode $E_{\text{ph}}(\theta)$ is related to the transmission angle

$$E_{\text{ph}}(\theta) = E_c * \left( \sqrt{1 - \sin^2 \theta / n_{\text{eff}}^2} \right)^{-1}$$

where the fitting parameters $E_c$ and $n_{\text{eff}}$ are the cavity photon energy at normal incidence and effective cavity refractive index, respectively. The values thus obtained for M-A cavity are $E_c = 2.34eV$, $n_{\text{eff}} = 1.54$. Relatively low effective refractive index confirms that most of the optical field is in the SiO$_2$ layers, with the modes matching well with the transfer matrix and two level model simulations [Fig. 3(B)]. Similar results are found for all-metallic (M-M) microcavities of optical length $7\lambda/4$. Here an additional 125nm buffer layer of polymer (PMMA) is spin-coated on the M-A cavity, and a final deposition of 10nm Ag ($R=35\%$) completes the microcavity. Once again, experimental angle-resolved transmission spectra match unambiguously with transfer matrix model simulations [9]. These Rabi splitting values of both M-A and M-M cavities are much larger than that of previously reported all-metallic organic (J-aggregate) microcavities, and comparable to hybrid microcavities of similar cavity lengths.

**4. CONCLUSIONS**

Here we fabricated and studied layered perovskite inorganic-organic multiple quantum well heterostructures. These narrow-and strong exciton-linewidth hybrids show little Stokes shift and are highly suitable for strong-coupling applications. Incorporating such hybrid thin films in M-A and M-M low-Q cavities provides room-temperature strong coupling phenomena. The Rabi splitting is found to be ~130meV for M-A and ~160meV for M-M low-Q microcavities respectively. Strong coupling of these IO-MQWs paves the way for new device applications, particularly optimal for polariton laser experiments, since these films are more robust in nonlinear experiments than comparable organics. Further, these hybrids are of significance in fabricating robust optoelectronic devices.

**ACKNOWLEDGEMENT**

Authors are thankful to Department of Science & Technology (DST), India for the financial support. This work is part of **UK-India Education Research Initiative (UKIERI)**.

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