

All-optical nonlinear phase modulation in open semiconductor microcavities

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We report a significant advancement in ultra low power light-by-light phase modulation using open semiconductor microcavities in the strong light-matter coupling regime. We achieve cross-phase modulation of up to 247 ± 17 mrad per particle between laser beams attenuated to single-photon average intensities. This breakthrough extends the potential for quantum information processing and nonlinear quantum optics in strongly coupled light-matter systems, setting a new benchmark in the field without relying on atom-like emitters. Our findings suggest promising new avenues for scalable quantum optical technologies.

Introduction

Strong effective interactions between photons, mediated by material nonlinear optical response, underpin solutions to a wide range of scientific and technological challenges from quantum computing and entanglement distribution [1–4], to the ultra-low power limit of classical all-optical processing [5], to novel approaches to quantum machine learning [6]. This has inspired great effort to build photonic systems exhibiting cross-phase modulation (XPM) where one optical signal can strongly and deterministically modulate the phase of a separate optical signal at the single-photon level. Such single photon phase shifters underpin routes to applications such as two-photon gates [7–11] for quantum information processing, photon number detectors and sorters [12], non-Gaussian state generation for metrology [13], programmable quantum circuits [14], and quantum optical neural networks [15].

Single photon phase shifts up to π have been observed using single [16, 17] or ensembles of natural atoms [18, 19]. For simplicity and scalability it is desirable to incorporate the nonlinear elements on semiconductor chips. Recently, semiconductor artificial atoms (quantum dots, QDs) [20, 21] have allowed XPM phase shifts of ~ 600 mrad [22] and have already been utilised for quantum simulation [14]. Challenges remain, however, with scaling up to circuits containing many dots with the same operating frequency.

An intrinsically more scalable approach is to couple light to two-dimensional excitons confined in quantum wells (QW), which have homogeneous energies on large (millimeter) spatial length scales. In the strong light-matter coupling regime this results in formation of part-photon part-exciton superposition states called exciton-polaritons. 0D spatial confinement can then be applied entirely through the photon degree of freedom using well

established photonic fabrications techniques. Interactions between the excitonic components of the polaritons are at least 1000 times stronger than in semiconductors in the weak light-matter coupling regime and several works have used them to manipulate quantum number statistics within single polaritonic modes [23–26].

A further advantage is that the nonlinear bandwidth, related to the light-matter coupling rate or vacuum Rabi splitting Ω_{Rabi} , exceeds several meV allowing ultrafast (picosecond) operation. The micrometer size of the optical modes makes coupling light in and out relatively simple. The tradeoff for these advantages is the smaller interaction strength compared to QDs. In a previous demonstration in solid microcavities [27] we showed XPM of 3 mrad per particle, limited by large cavity size, low excitonic content of the polaritons, and broadening of polariton linewidth due to etching through the QW. Despite these limitations the result was already an order of magnitude higher than any other systems not relying on atom-like emitters.

In this work, we experimentally study XPM in a tunable open-access polariton microcavity [28] with significantly tighter spatial confinement (beam waist $1\ \mu\text{m}$) and narrower excitonic resonances allowing smaller polariton linewidths ($\sim 50\ \mu\text{eV}$) at the higher excitonic content (70%) where interactions are maximised. By these means we significantly advance the state of the art single polariton XPM by a factor of 80, up to 247 ± 17 mrad. We furthermore show modulation of a probe by 500 mrad (50%) using only 10 control particles, corresponding to a peak intra-cavity energy of only 2 aJ. We use our highly flexible platform to study time-delay, frequency detuning, polarisation and power dependences and address the microscopic physics underlying these giant interactions. We find that there is negligible contribution from long-lived (> 300 ps) excitonic species. Furthermore, we find that the strength of the nonlinear response varies rapidly with frequency and becomes negligible for negative cavity-exciton detunings. This trend cannot be explained by the frequency dependence of excitonic fraction and linewidth usual for polaritons and suggests an

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additional microscopic mechanism underlying the nonlinearity.

Sample design and fabrication

Our polaritonic system is based on an open-access microcavity [28] formed between a dielectric concave distributed Bragg reflector (DBRs) and a planar semiconductor sample. The latter contains a 17 nm wide InGaAs quantum well (QW) in a GaAs spacer layer above a GaAs/AlAs DBR. The dielectric and semiconductor components are separated by an air gap, with their distance controlled by piezo positioner stages. Such hemispherical cavities support a lowest order mode with a tightly confined beam waist close to the quantum well position [28]. To minimise absorptive losses in the semiconductor material the whole cavity is kept at a temperature of 4 K by a helium exchange gas coupled to a bath of liquid helium.

Figure 1b shows a white light transmission spectrum as the separation between the mirrors is varied, revealing clear anti-crossing between lower polariton branch (LPB) and upper polariton branch (UPB) on either side of the exciton (X^0). The vacuum Rabi splitting $\Omega_{\text{Rabi}} = 2.87$ meV between the two branches characterizes the light-matter coupling strength. The excitonic (photonic)

content of the polaritons $|X|^2$ ($|C|^2 = 1 - |X|^2$) increases (decreases) as the lower polariton energy approaches the exciton resonance.

Figure 1c presents the lower polariton linewidth extracted from white light transmission spectra at each detuning (blue curve). The orange curve is a fit of the model $\gamma = |C|^2 \gamma_C + |X|^2 \gamma_X$ to the data where γ and γ_C and γ_X are the polariton, photon and homogeneous exciton linewidths respectively. The linewidth reduces significantly from $\gamma_C = 113 \pm 9 \mu\text{eV}$ in the photonic regime to $\sim 50 \mu\text{eV}$ at high exciton fraction $|X|^2 = 54\%$. The decreasing linewidth at high $|X|^2$ indicates negligible contribution of the exciton inhomogeneous broadening to the linewidth for these detunings. This is consistent with previous measurements on samples with similar 17 nm wide InGaAs QWs [29] and can be explained by the well-known phenomenon of motional narrowing [30–32]. The ability to reach narrow linewidths dominated by the exciton homogeneous linewidth for highly excitonic (and therefore nonlinear) polaritons is one of the key factors in our achieving high single photon phase shifts. For several cavity-exciton detunings between -7 and 0 meV we also confirmed the linewidth by scanning the cavity mode energy through a fixed single-frequency laser line and observing the Lorentzian shaped transmission vs. energy peaks. The measured linewidths agree with those from the white light transmission within the uncertainty. We note that the cavity also exhibits a birefringent splitting between two linearly polarised eigenstates which likely arises due to strain in the semiconductor sample [33]. The splitting between the polarisation states in the purely photonic limit is $97 \pm 12 \mu\text{eV}$.

Experiment

The phase rotation measurement is illustrated in Figure 1a. We generate pump and probe pulses using electronically triggered high-speed electro-optical modulators (EOMs) to carve single-frequency continuous wave lasers into pulses. This approach allowed precise control over the repetition rate, pump-probe delay, and pulse width ranging from approximately 50 ps (probe) to 1.3 ns (control). The incident pulses were tuned in frequency to resonantly excite the lowest lying transverse cavity mode. The control (pump) and signal (probe) beams had the same frequency (corresponding to the maximum transmission) but differing pulse temporal envelopes and were derived from separate lasers so that they had a random relative phase. The pulses were attenuated to produce peak intra-cavity photon numbers in the range 1-30. The signal beam was linearly polarised with polarisation aligned to one of the birefringent split polarisation states (horizontally (H) polarised in the laboratory frame at the sample position). The incident control beam polarisation could be tuned continuously between left and right cir-

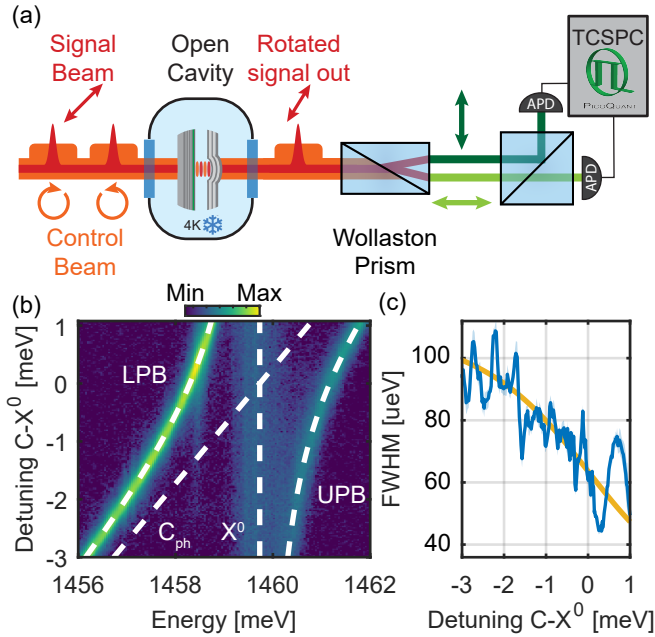


FIG. 1. (a) Simplified schematic of the experimental setup. (b) White light transmission spectra as a function of the detuning between bare cavity mode (C_{ph}) and exciton (X^0) plotted in logarithmic colour scale. (c) Polariton linewidth extracted from the white light transmission measurements, as a function of the detunings of the cavity mode from the exciton resonance (blue curve) and calculated from the simulation (orange curve).

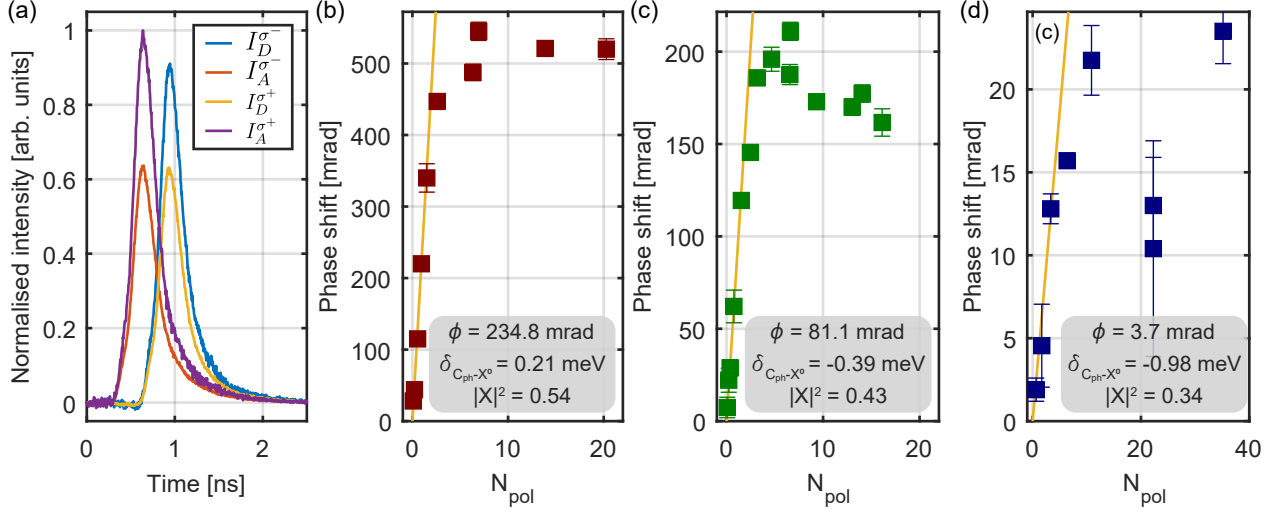


FIG. 2. (a) Example TCSPC traces recorded during a phase-shift measurement. The control-beam background has been subtracted. A and D denote the avalanche-photodiode detection channels measuring the two orthogonal polarisation components. σ^+ and σ^- indicate the control-beam polarisation state. (b–d) Measured phase shift as a function of the control beam’s mean polariton number for different cavity–exciton detunings.

cular, passing through H linear polarisation, by means of a free-space EOM acting as a variable waveplate.

The polaritonic interactions in the cavity cause the control pulses to rotate the probe linear polarisation state away from H [27]. The change in linear polarisation direction corresponds to a phase shift ϕ between circular polarisation components. After being transmitted through the cavity the output polarisation state is resolved in the diagonal (D) / anti-diagonal (A) basis (at the sample position) by a half wave plate and Wollaston prism polarizer allowing maximum sensitivity to changes in the probe polarisation direction.

The transmitted photons were detected by single photon avalanche diodes (SPADs) and sorted into time bins using time-correlated single photon counting (TCSPC) electronics (PicoQuant HydraHarp) synchronised to the laser pulse triggering. This enabled us to record the pulse temporal envelopes with high signal to noise ratio and high temporal resolution and thus separate the pump and probe by temporal shape. Measurements were taken with the probe beam both on and off and the reference control pulse shape was subtracted to leave only the probe pulses. External marker channel inputs to the TCSPC electronics also allowed photon counts to be binned according to control beam polarisation state allowing high speed (kHz) repetitive polarization dependence sweeps.

Figure 2(a) shows an example of the recorded probe pulse temporal profiles (convolved with the SPAD impulse response). $I_{D,A}^{\sigma^+,\sigma^-}$ are the intensities for diagonal (D) and anti-diagonal (A) components for σ^+ and σ^- polarised control beams. Without the control beam present

the H polarised probe results in equal count rates on the SPADs measuring the D and A polarization components. The phase shift ϕ can be deduced from the change in the count rates and in particular by the diagonal polarisation degree (S_2 Stokes parameter) [27]. Furthermore, control beams with opposite circular polarisation lead to ϕ with opposite sign (which we will examine in more detail later). The phase shift is then given by

$$\phi \approx \frac{1}{2} \left(\frac{I_D^{\sigma^+} - I_A^{\sigma^+}}{I_D^{\sigma^+} + I_A^{\sigma^+}} - \frac{I_D^{\sigma^-} - I_A^{\sigma^-}}{I_D^{\sigma^-} + I_A^{\sigma^-}} \right) \quad (1)$$

Here $I_{D,A}^{\sigma^+,\sigma^-}$ are sums of the curves in Fig. 2(a) over a small range of times near the peak of each pulse.

Results

One of the main advantages of using an open cavity system for studying polariton systems is that this setup is tunable, allowing us to measure the phase shift across a broad range of parameters, specifically for states with varying excitonic fractions. Figures 2 b-d display the power dependencies of the phase shift for three detunings of the cavity mode from the exciton resonance. All presented results demonstrate similar behaviour: the phase shift increases linearly with the number of polaritons in the cavity and saturates after reaching 5-10 polaritons. The value of the phase shift per polariton was extracted as the slope of the linear fit to the linear part of the graph for each detuning. As a result, we observed phase shifts ranging from 3.7 mrad to 234.8 mrad per polariton for polariton states with different excitonic fractions

from 34% to 54%.

Figure 3a shows the variation in phase shift for 8 measurements over a wide range of detunings. The highest phase shift observed was 247 ± 17 mrad per polariton for an excitonic fraction of 55%. These results indicate a 80x times enhancement in phase shift per polariton compared to that reported in a solid micropillar [27]. Moreover, the present system allows observation of an absolute phase shift of 500 mrad (50%) with only 10 control polaritons in the cavity, see Fig. 2b. These 10 particles correspond to an intra-cavity energy of ~ 2 aJ. This absolute phase shift is more than 30 times higher than that demonstrated in polariton micropillars, where the phase shift was limited to 15 mrad by saturation [27].

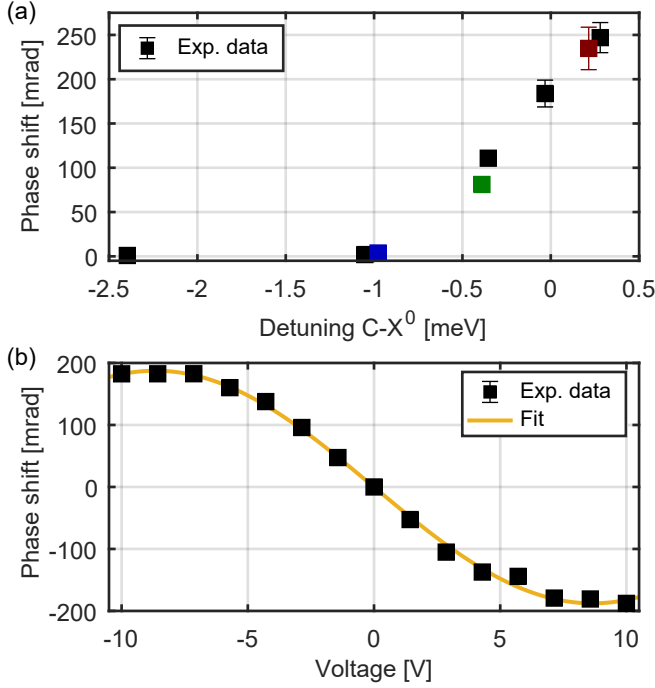


FIG. 3. (a) Phase shift per 1 polariton as a function of cavity-exciton detuning. (b) Dependence of phase shift on applied voltage of the EO amplitude modulator. The experimental data (black squares) are fitted with a sine function (orange line).

We also confirmed that the phase shift had the dependence on control beam polarisation expected from previous works [27]. As described above the control polarisation was repeatedly scanned through 15 points every 7.5 ms allowing a parameter sweep immune from gradual experimental drifts. We performed polarisation sweeps for 4 detunings in the range from -1.05 meV to 0.28 meV for ≤ 1.5 control polaritons. Figure 3b shows a typical result obtained at a detuning of 0.28 meV and cavity occupancy of 0.8 control polaritons. All of the polarisation sweeps exhibited the same qualitative trend. The phase shift changes sign when the control beam polarisation is reversed (comparing the left- and right-most points),

is zero when the control polarisation is the same as the probe polarisation (middle point), and has the expected sinusoidal dependence on EOM voltage.

We now consider the dependence of the phase shift on the cavity-exciton detuning. For detunings below -0.5 meV the phase shift reduces markedly, reaching a value of only 3.8 mrad at a detuning of -1 meV and remaining low for more negative detunings. Comparing to the highest 5 phase shift points and accounting for the $|X|^4$ dependence of the interaction strength on detuning [27] as well as the measured frequency dependence of the polariton linewidth we expect phase shifts of 58 ± 13 mrad at this detuning where $|X|^2 = 0.34$ and $\gamma = 80$ μ eV. This is 15x higher than the measured phase shift. We note that the $|C||X|^3$ scaling of polariton nonlinearities originating from oscillator strength saturation would result in an even less steep expected frequency dependence and hence a larger discrepancy between expected and measured values. The rapid decrease in phase shift with detuning cannot be explained by the usual frequency dependence of polariton nonlinearity due to reducing excitonic fraction. The explanation requires some additional strong frequency dependence of the polariton interactions.

A possible origin for this strong frequency dependence is coupling of the polariton states to a biexciton resonance [34]. This mechanism has been well studied in polariton cavities and was recently used to explain the frequency dependence of nonlinear polariton number correlations in cavity structures very similar to ours [25, 26]. It should be noted that biexcitons cannot be generated using co-circularly polarised beams. However, in our experiment, the use of a linearly polarised probe beam ensures the presence of cross-polarised polaritons. The biexciton coupling leads to a Feshbach-like resonance when the polariton energy is equal to half the biexciton energy. For biexciton binding energies in the range 2.2 to 3.0 meV [25, 34] the resonance is expected to occur for polaritons with energy 1.1 to 1.5 meV below the exciton. With our Rabi splitting 2.87 meV these correspond to cavity-exciton detunings -0.12 to $+0.77$ meV which approximately corresponds to the spectral region where we observe strong nonlinearity. However, further analysis is required to confirm or rule out this mechanism as the origin of the observed rapid frequency dependence of the nonlinear response.

As we generate the control and probe pulses using electronically triggered modulation of CW lasers this setup permits precise control over the pulse duration, repetition rates, and temporal delays. We utilised this system to measure the response time of the optical nonlinearity underlying the XPM. Figure 4a displays the measured phase shift versus time delay while Fig. 4b shows three different delays between control and probe pulses. For an instantaneous nonlinearity the phase shift vs. delay is expected to be the same shape as the control pulse. A number of studies have demonstrated that polariton non-

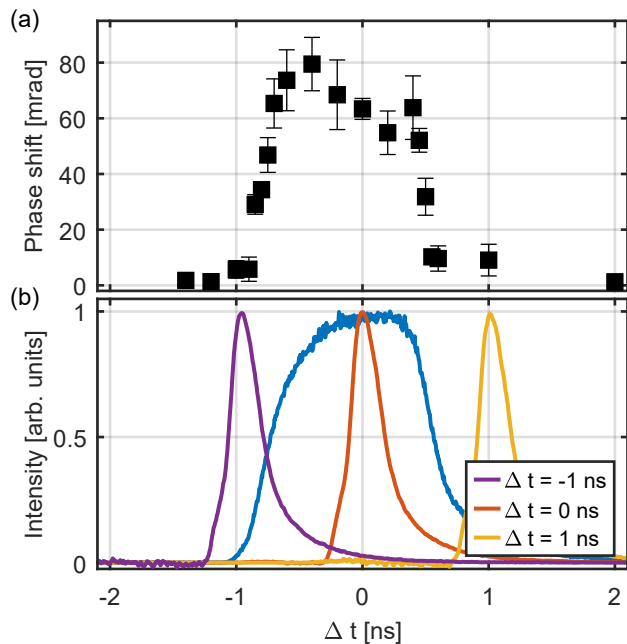


FIG. 4. (a) Phase shift dependence on time delay between probe and control pulses. (b) Control pulse (blue) and probe pulse for three different delays. Note that the pulse shapes are convolved with the timing jitter of the SPADs.

linearities can be affected by a reservoir of dark exciton states, even for resonant excitation. The occupation of these states builds up and decays on timescales of a few hundred picoseconds [35–37]. For nonlinearities dominated by long-timescale effects the phase shift vs. time is expected to exhibit an exponential rise and/or fall on the leading/trailing edge corresponding to build-up and/or decay of the reservoir. In our case, the rise and fall times (between 10% and 90% of the peak of the phase shift) are 290 ps and 210 ps, respectively. These agree within 15 ps with the rise and fall times of the control pulse intensity measured independently using a streak camera.

This indicates that the nonlinearity causing the phase shift does not originate from reservoirs with timescales long compared to ~ 300 ps. This is consistent with recent observation of the Bogoliubov excitation spectra of polariton condensates in cavities with very similar QWs, which have also shown negligible influence from reservoir effects [29].

CONCLUSION

In conclusion, our research has demonstrated all-optical phase shift in an open cavity polariton system, achieving up to 247 ± 17 mrad per polariton and setting a record for the highest observed value without atom-like emitters. Furthermore, we achieve an absolute phase shift of 500 mrad, surpassing previous achievements in

polariton devices by 35 times. These results not only underscore the potential of polariton systems in nonlinear quantum optics but also highlight the scalability and efficacy of open cavities for future quantum information processing applications. This study marks a significant step forward, opening up novel possibilities for the development of quantum optical devices based on polariton interactions.

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