Few-photon all-optical phase rotation in a quantum-well micropillar cavity

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Photonic platforms are an excellent setting for quantum technologies as weak photon–environment coupling ensures long coherence times. The second key ingredient for quantum photonics is interactions between photons, which can be provided by optical nonlinearities in the form of cross-phase modulation. This approach underpins many proposed applications in quantum optics3,5 and information processing7, but achieving its potential requires strong single-photon-level nonlinear phase shifts as well as scalable nonlinear elements. In this work we show that the required nonlinearity can be provided by exciton–polaritons in micropillars with embedded quantum wells. These combine the strong interactions of excitons16–18 with the scalability of micrometre-sized emitters19. We observe cross-phase modulation of up to 3 ± 1 mrad per polariton using laser beams attenuated to below the average intensity of a single photon. With our work serving as a stepping stone, we lay down a route for quantum information processing in polaritonic lattices.

Quantum applications of cross-phase modulation (XPM) include teleportation1, photon-number detection2, metrology3, cryptography4 and quantum information processing (QIP), where XPM was proposed as a route to circuit5 and measurement-based6 quantum computing; however, XPM-based photonic QIP faces several challenges. Frequency entanglement can degrade the fidelity12 of quantum gates3, but this can be overcome by cascading nonlinear resonators, with each providing a moderate phase shift. Such cascading naturally requires scalability of the resonators. The remaining major challenge, which we address in this paper, is to find a system with high enough single-particle XPM phase shift, ϕsp, which is suitable for scaling.

The small size of atom-like emitters ensures strong interactions and large ϕsp, but at the same time makes scalability challenging. Real atoms are not easy to trap and manipulate, and it is difficult to achieve many solid-state artificial atoms with deterministic frequencies and locations. Experimentally, ϕsp from 0.1 to π radians have been observed in atomic ensembles13,14 and atoms15 or quantum dots16–18 strongly coupled to cavities. Methods using electro-magnetically induced transparency15–16 or sequential photon–atom interactions17,18 achieve ϕsp = π using microsecond pulse sequences and milliwatt ancillary beams, whereas passive nonlinear XPM, as we study here, is favoured for high-rate, low-energy operation.

Approaches avoiding atom-like emitters have been hindered by the small optical nonlinearity in typical Kerr media; ϕsp values from 10−4 to 0.3 mrad have been demonstrated with optical fibre19 and atomic vapours20,21. Polariton micropillars, where photons are strongly coupled to excitons22, are a prime candidate for combining high phase shifts and scalability. Their micrometre dimensions allow scaling into large lattices with deterministic positioning and energies identical within the linewidth23, whereas the excitonic component of polaritons provides interactions at least 1,000-times larger than in weakly coupled and/or bulk semiconductors24. An important feature of polariton interactions is their polarization dependence25, which can be used to implement all-optical spin switches26 or break time-reversal symmetry. Polaritonic resonators have been used as a source of weakly sub-Poissonian light27. However, neither XPM between distinct modes nor the polarization dependence of interactions has been harnessed at the few-particle level.

In this article we demonstrate ϕsp values of up to 3 mrad, the highest amount without using atom-like emitters. As a proof of principle, we demonstrate this phase rotation using a control laser attenuated down to an average intensity of 0.13 polaritons, where the probability of >1 control polariton being in the system is <1%. We exploit the polarization-dependent interactions to encode the XPM on the polarization state of a second laser, achieving high phase sensitivity and stability. Extrapolating our experimental results to samples with tighter photon confinement and narrower exciton linewidth28,29, we predict ϕsp approaching a substantial fraction of π. Using the example of XPM-based conditional-phase (CPHASE) quantum gates, we show theoretically that these experimental results open new routes towards active quantum processing with exciton polaritons.

Our device is an AlGaAs air-post Fabry–Pérot microcavity containing a single quantum well of the type illustrated in Fig. 1a (see Methods and Supplementary Discussion 1). All experiments were performed near liquid helium temperature. We first characterized the micropillar using photoluminescence spectroscopy. The spectrum of discrete states resulting from the three-dimensional optical confinement can be seen in Fig. 1b, with mode intensity profiles shown in Fig. 1c.

The phase rotation measurement is illustrated in Fig. 1a (see also Supplementary Discussion 1). We resonantly excited E12 with a circularly polarized continuous-wave beam (control beam) and E13 with a pulsed beam linearly polarized along the y direction.

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Fig. 1 | Sample properties. a, Schematic of the experimental arrangement. \(k_y\) is the \(y\) component of the wavevector and \(k_\parallel\) is the non-zero peak wavenumber of the signal state. b, Photoluminescence spectrum from pillar A; \(\hbar \omega_{01}=1.446\) meV. c-e, Real space photoluminescence intensity maps taken at the frequencies of the ground (\(E_{11}\)) (c) and first excited (\(E_2\) (d) and \(E_3\) (e)) manifolds. The scale bar corresponds to 5 μm, which is the size of the micropillar. Colour scale is linear and denotes intensity relative to peak. f, Example of raw TCSPC curves recorded during a phase shift measurement; |\(D\rangle\) and |\(A\rangle\) denotes the signals from avalanche photodiodes (APDs) measuring those polarization components while ‘Ctrl on’ and ‘Ctrl off’ specify the control beam state. The inset to f shows a zoom of the region labelled by a black rectangle.

(signal beam). The signal linear polarization can be decomposed into two circularly polarized components. As polariton interactions depend strongly on relative circular polarization\(^\text{17}\), the presence of the control beam shifts the \(E_{11}\) resonance to higher energies only for the polarization parallel to the control beam. Consequently, the co-polarized signal component acquires a relative phase shift via XPM, resulting in a rotation of the signal beam linear polarization angle. Measuring this change in polarization reveals the amount of phase shift. A quantitative analysis of the XPM and detection process is given in Supplementary Discussion 2. The overall nonlinear phase shift \(\phi\) reads

\[
\phi = \frac{2}{\gamma/2} |X_{11}|^2 |X_{12}|^2 N_{pol} \frac{A_{\text{int}}}{A_{\text{eff}}}.
\]

Here, \(N_{pol}\) is the mean number of control polaritons present in the cavity, \(A_{\text{int}}\) is the averaged confinement area of the modes, and \(\gamma\) is the full width at half maximum (FWHM) signal linewidth; \(g_1\) and \(g_2\) are the interaction strengths for co- and cross-circularly-polarized excitons, respectively. The polariton interaction strength increases with the excitonic fractions |\(X_{11}\)|\(^2\) and |\(X_{12}\)|\(^2\) of the control and signal states. The nonlinear frequency splitting between circular-polarization states is analogous to a Zeeman splitting caused by an effective magnetic field and the polarization rotation is analogous to the Faraday effect.

To measure the XPM phase shift we collected the transmitted light, filtered out the control beam using a spectrometer, and measured the diagonal (|\(D\rangle\)) and anti-diagonal (|\(A\rangle\)) signal polarization components with the control beam alternated between on and off. Intensities were measured using time-correlated single-photon counting (TCSPC), which allowed further separation of the pulsed signal beam from the continuous-wave control (see Methods and Supplementary Discussion 3). An example of the TCSPC data is shown in Fig. 1f. The peaks are due to the signal pulses, whereas effects uncorrelated with the signal pulses form a continuous-wave background, which we measured using points at times far from the peak and subtracted. We then integrated the counts around the peaks to obtain the total signal count rates \(I_{\text{on}}^{\text{D}}, I_{\text{on}}^{\text{A}}, I_{\text{off}}^{\text{D}}, I_{\text{off}}^{\text{A}}\) for |\(D\rangle\) and |\(A\rangle\).

The phase shift \(\phi\) is deduced from the difference in polarization degree with the control beam on and off (see Supplementary Discussion 2) and, for small nonlinear resonance shifts compared with the linewidth, is given by

\[
\phi \approx \frac{I_{\text{on}}^{\text{D}} - I_{\text{off}}^{\text{D}}}{I_{\text{on}}^{\text{D}} + I_{\text{off}}^{\text{D}}} - \frac{I_{\text{on}}^{\text{A}} - I_{\text{off}}^{\text{A}}}{I_{\text{on}}^{\text{A}} + I_{\text{off}}^{\text{A}}}.
\]

As well as measuring the phase, it is important to accurately deduce \(N_{pol}\). The absolute calibration of which was obtained by measurement of the cavity transfer function, carefully separating the radiative losses from other contributions to the linewidth, which leads to

\[
E_{\text{cav}} = \hbar \omega_{01} N_{pol} = \frac{P_{\text{out}}}{\gamma T |\langle C_{11}\rangle|^2}.
\]
The phase shift increases with increasing fractions and linewidths (see Methods). The phase shift increases for the transmitted power and $g_e$ lower end of the range established by many groups\(^9,10,26,33–36\), indicating that we do not underestimate $N_{\text{pol}}$ of reservoir states using samples with smaller exciton inhomogeneous broadening\(^32\). We note that saturation will not be detrimental to performance as devices will operate with independent of control polarization and has a weak dependence on control power arising from blue-shift of the states relative to resonance with the signal laser (Methods). To maximize transmission, low sample temperatures of 4 K (easily achieved using commercial closed-cycle cryostats) are important, in comparison to refs. \(^13,14,15,24\), where room temperature phase shifts were demonstrated.

**Table 1** | Comparison of XPM phase shift platforms

| System                      | $\phi$ per particle (mrad) |
|-----------------------------|-----------------------------|
| Rydberg atoms in EIT regime\(^7\) | 3,300                       |
| Single caesium atom\(^13\)  | 280                         |
| Strongly coupled quantum dot\(^22\) | 220                          |
| This work                   | $3 \pm 1$                   |
| Rubidium vapour in a hollow-core fibre\(^4\) | 0.3                         |
| Metastable xenon\(^15\)     | 0.0003                      |
| Photonic crystal fibre\(^23\) | 0.0001                      |

At above ~6 polaritons (Fig. 2b,c), the phase shift saturates for both pillars. Further experiments are needed to identify the mechanism behind this saturation. The dependence on control polarization is preserved even up to 42 polaritons (Fig. 3a), but with reduced magnitude, which suggests that saturation is due to suppression of the polarization dependence of the effective polariton nonlinearity, or of the control polariton circular polarization degree. These can occur due to interaction with a reservoir of excitons, which can be enhanced by sample heating\(^29,31\). Reservoir accumulation and thermal effects can be overcome using pulsed rather than continuous-wave control excitation\(^32,30\), and by reducing the density of reservoir states using samples with smaller exciton inhomogeneous broadening\(^22\). We note that saturation will not be detrimental in experiments with very low $N_{\text{pol}}$ of 0.13 ± 0.03. The data in Fig. 2a and Fig. 2b give phase shifts of 0.04 ± 0.06 and 0.08 ± 0.05 mrad, respectively. The four points in Fig. 3c produce phase shifts larger than the uncertainty given by the error bars. We are thus able to measure a phase shift for average powers $P_{\text{pol}} = 0.13$, where the probability of an occupancy greater than 1 is less than 0.8%, based on the Poissonian statistics expected for laser fields. We are thus well inside the single-photon regime.

In Table 1 we compare the phase shifts of various systems. Our phase shift of 3 mrad per polariton in pillar B is consistent with a blue-shift of only 0.062 μeV per polariton compared to the $|E_1\rangle$ state linewidth 83 μeV, which highlights the sensitivity of the technique.

For our proposed Faraday-like phase rotation mechanism the induced phase should follow a sinusoidal dependence on the angle of the quarter-wave plate used to set the control polarization, vanishing when the control is linearly polarized and reversing sign when it is switched to the opposite circular polarization (see Supplementary Discussion 2). In Fig. 3 we show the phase shift versus the quarter-wave plate angle for three different control beam strengths covering two orders of magnitude. It agrees well with the theoretical prediction for all control powers, reducing to zero at around 90° and then reversing sign. We note that the absolute magnitude of the phase shifts is different to those in Fig. 2 due to day-to-day drifts in sensitivity (see Methods).

We measure control (signal) beam transmissions of 45% (35%) in our current devices, limited by a combination of imperfect mode-matching and absorptive losses (see Supplementary Discussion 8 for more details). Transmission can realistically be increased above 90% using lower temperature ~4 K and GaAs quantum wells, which have lower inhomogeneous broadening\(^32\). This would allow transmission through multiple pillars with a $1/e$ decay of approximately ten sites. The signal transmission is independent of control polarization and has a weak dependence on control power arising from blue-shift of the states relative to resonance with the signal laser (Methods). To maximize transmission, low sample temperatures of 4 K (easily achieved using commercial closed-cycle cryostats) are important, in comparison to refs. \(^13,14,15,24\), where room temperature phase shifts were demonstrated.
Having demonstrated XPM for single-polariton intensities it is interesting to consider whether XPM-based effects can be used for polaritonic QIP. Entanglement between frequency states was shown to limit the fidelity of XPM-based CPHASE gates for large phase shifts. This obstacle can be overcome if nonlinearity is distributed over several cavities with cascaded wavepacket propagation. In Supplementary Discussion 9 we analyse theoretically a potential scheme to achieve this. It requires scattering through order ten resonators and optical circulation (suppressed backscattering). Micropillar lattices of this scale are regularly produced, whereas directional propagation from one phase-shift element to the next can be achieved using the edge states in polariton topological insulator lattices and benefit from the rich topological physics of polaritons.

As we show in Supplementary Fig. 10, CPHASE gate fidelity depends on the size of the single-polaron phase shifts and the target phase. Full π shift, corresponding to high-fidelity controlled-Z gate, requires large single-polaron nonlinearity-to-linewidth ratio $U_{pp}/\gamma$; however, near-perfect fidelity $\pi/m$ gates (where $m$ is an integer) can even be achieved at $U_{pp}/\gamma < 1$. When concatenated, these contribute to a universal gate set, and, at $m = 6$, have been used in quantum hardware-based solution of optimization problems.

Although a distant goal, we consider the ability to inject nonlinearity at the single polariton level a crucial element for many QIP protocols. In general terms, our quantum well polariton approach to single photon phase shifts provides the tools to optimize the balance between scalability and interaction strength for any given application. In summary, we demonstrated a few-particle polariton XPM phase shift in a scalable on-chip platform. This opens up new approaches to a wide class of nonlinear quantum optical phenomena, and offers a route towards QIP with polaritonic lattices.

Online content

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Methods

Sample properties. The sample consists of a GaAs cavity containing a single 15-nm-wide In_{0.05}Ga_{0.95}As quantum well at the electric field antinode and embedded between two Al_{0.1}Ga_{0.9}As/Al_{0.95}Ga_{0.05}As Bragg mirrors. Confinement of the light in all three dimensions results in discrete optical modes, in the cavity, which were measured using imaging photoluminescence spectroscopy (Fig. 1b–e). The transverse mode profiles are similar to Hermite–Gauss modes. In the ground state manifold, labelled $E_0$, there are two degenerate polarization states. In the first excited manifold the sub-manifolds $E_1$ and $E_2$, have non-zero wavevector component in the $y$ and $x$ direction respectively and each contains two orthogonal polarization states. There is a small splitting among these four states due to a combination of TE–TM splitting and the pillars not being perfectly square. The splitting between the $E_1$ and $E_0$ submanifolds allowed them to be mapped separately (Fig. 1d,e) using energies one FWHM either side of the peak (see Methods).

The studied sample contains many pillars with different sizes (2–5 μm width) to allow tuning of the spatial distribution of modes and their energy separation. The spacing between individual pillars was 10 μm. Different detunings of pillar modes with respect to the quantum-well excitation resonance were also available due to a wedge in the molecular-beam-epitaxy grown cavity. The experiments presented in this paper were performed on 5 μm x 5 μm square pillars. The square geometry allowed simple excitation of the Hermite–Gauss-like first excited state compared to more difficult beam shaping required to excite the Laguerre–Gauss-like excited states of a circular pillar. The 5 μm size of the pillars minimized TE–TM splitting of the first excited manifold. The TE–TM splitting of the $E_1$ mode was obtained from a coupled excitation-photon oscillator model fit to the dispersion of pillar modes. To determine the detuning of the modes from the exciton we compared the energy splitting between the $E_1$ and $E_0$ modes with the value for very negatively detuned pillars. The splitting reduces as the photonic fraction reduces and so can be used to directly obtain the photonic (and hence excitonic) fraction of the polaritons.

Number of polaritons. The number of control polaritons in the pillar $N_{\sigma}$ was deduced using the transmitted power and the radiative loss rate through the mirror on the transmission side of the sample, $\gamma$. The accuracy of equation (3) was confirmed by comparison of transmitted power and stored electromagnetical energy using exact solutions of Maxwell’s equations (transfer matrix method) for cavities with a wide range of parameters around those of the experimental device (see Supplementary Discussion 5).

Common experimental details. Experiments were performed near to liquid helium temperature. The sample was held in vacuum and mounted to a copper block connected to the heat-exchanger of a continuous flow cryostat. The copper block was held at less than 5 K, as measured using a silicon diode temperature sensor. The radiation through the cryostat windows and the small transverse area for heat flow in a 5 μm size micropillar may have caused the actual pillar temperatures to be higher.

The micropillars were optically excited directly (not through the substrate) using a 4 mm focal length objective (numerical aperture 0.42). Light emitted by or transmitted through the pillars was collected by a 10 mm focal length microscope objective (numerical aperture 0.6) and imaged via a set of confocal lenses onto the entrance slit of an imaging spectrometer. The spectrometer output could be switched between a CCD camera and an exit slit used to select only the signal beam for the APD measurements. The spectrometer exit slit was imaged onto the APDs via another pair of confocal lenses.

Phase rotation measurement details. For the phase rotation experiments, we resonantly excited the micropillar ground state with a circularly polarized control beam emitted by a continuous-wave single-mode laser. At the same time, we also excited the $E_1$ pillar mode with a linearly polarized signal beam from a tunable mode-locked titanium:sapphire laser with a pulse duration of ~100 ps and a repetition rate of 80 MHz. The sizes and divergences of the input control and signal beams were controlled with telescopes to match those of the pillar modes and hence optimally couple light to the microcavity. Both the signal and control beam were set to have a flat-phase beam waist of ~3 μm (FWHM) on the sample surface, matching the ground-mode FWHM. After this, a phase mask was placed at the focus of the telescope controlling the signal beam to introduce a π phase jump at the centre of the signal spot on the sample surface. In this way, the signal beam was converted to a Hermite–Gauss-like beam with symmetry matching that of the $E_1$ mode but of the wrong symmetry to excite the $E_0$ mode. To ensure that experimental drifts did not compromise optimal coupling the transmission of the control and signal beams was checked after every data point shown in Figs. 2 and 3 and reoptimized if necessary.

Measurement of the intensities of the two polarization components was performed by photon counting using APDs owing to their extremely small noise level. The control beam was chopped on and off at a rate of 10 kHz using an electro-optic modulator driven by a square wave control signal. Counts from the APDs were sent to a TCSPC card via a router that encoded information about which APD detected the photon, and whether the chopped control beam was on or off. The signal beam was attenuated such that on average 0.025 photons were detected per laser pulse. We avoided spurious signals in several ways: (i) A signal beam was chopped in continuous wave whereas the signal was pulsed; (ii) we were able to remove any potential scattered control light reaching the APDs by subtracting the continuous wave background from the data. This also removed any dark or APD after-pulsing counts. The true signal counts were then obtained by summing counts around the signal peaks in the TCSPC traces. As we measure a polarization degree of the form shown in equation (2) any overall drifts or jitter in signal beam transmission or integration time simply cancel out. Our chopping of the control beam at 10kHz eliminates any control drift effects in a manner similar to lock-in detection, while collection over several minutes effectively averages out control beam jitter. Further details are given in Supplementary Discussion 3.

Number of polaritons. The number of control polaritons in the pillar $N_{\sigma}$ was deduced using the transmitted power and the radiative loss rate through the mirror on the transmission side of the sample, $\gamma$. The accuracy of equation (3) was confirmed by comparison of transmitted power and stored electromagnetical energy using exact solutions of Maxwell’s equations (transfer matrix method) for cavities with a wide range of parameters around those of the experimental device (see Supplementary Discussion 5).

The total radiative loss through both mirrors was obtained by measuring the linewidth $\gamma_{\text{total}} = 25.5 \pm 1 \text{ μeV}$ at a very photonic detuning where the losses are dominated by the finite reflectivity of the mirrors. It agrees well with transfer matrix simulations. We then use $\gamma = \gamma_{\text{total}} - \gamma_{\text{loss}}$, where $\gamma_{\text{loss}} = 0.553$ is related to the relative mirror reflectivity and was calculated using the transfer matrix method.

In principle either the incident or transmitted power can be used to obtain $N_{\sigma}$. We obtain a high transmission through the pillar with transmitted/incident power being 40% (45%) for the control state of pillar A (B). It is more accurate to use the transmitted power since incident power can be reflected due to imperfect mode matching.

The signal state transmission was 35% for the best coupling to the pillars that we achieved. The laser pulses incident on the pillar contained 27 photons on average. The transmitted pulses contained between 3 and 9 photons depending on coupling, corresponding to peak internal number of polaritons between 2 and 5 for pillar A, 3 and 8 for pillar B. We did not observe any dependence of the results on the number of signal polaritons.

The dependence of total signal beam transmission on the control was obtained by adding the intensities of both APDs (See Supplementary Discussion 8). We found no systematic dependence of the signal transmission on control beam polarization. The signal transmission versus control power varied with gradient sign. To check this further we performed a few measurements with a DC magnetic field applied with either positive or negative sign, depending on the dataset. This is consistent with a blue-shift of the states shifting further into or out of resonance with the signal laser (see Supplementary Discussion 2).

Statistical analysis. By calculating the mean and standard deviation $\sigma$ among many (10–100) repeated measurements of $\phi$, we directly obtain the average phase change and its uncertainty for a given beam. For $N_{\sigma}$, we use the standard error and the error bars are plotted covering the range from $-2\sigma$ to $+2\sigma$, which corresponds to the 95% confidence interval for a normal distribution.

Sources of noise in the data. There are two categories of noise contributing to the data shown in Figs. 2 and 3. These are a random error in the phase of individual data points, and systematic variations in sensitivity which occurred between individual data points and different data sets (see Supplementary
Discussion 3). The former arises from the Poissonian counting statistics. The latter arises because sub-linewidth changes in signal beam detuning can change the sensitivity of the measurement to the blue-shift of the states. The small changes in detuning were caused by slow frequency drift during data collection, necessitating slight retuning between data points, and also by the coarse tuning (limited to \( \pm 20-30 \mu \text{eV} \) accuracy) of our 100 ps pulsed laser. The sensitivity function is Lorentzian with the state linewidth (see Supplementary Discussion 2). During data collection, small experimental drifts were corrected in between recording each data point, resulting in small changes in sensitivity and hence some point-to-point noise. Nevertheless, the overall trends are clearly visible in the curves presented in Figs. 2 and 3 and they agree well with theory so we can be confident that this point-to-point noise is not too large. The dependence of sensitivity on small changes in signal laser detuning also causes small differences in scaling from one data set to another, hence the best-fit peak phase shift for \( N_{pol} = 42 \) polaritons in Fig. 3a is \( 5 \pm 1 \) mrad, which is slightly larger than the maximum value in Fig. 2b (\( 3.3 \pm 0.1 \)), which was measured on a different day.

Data availability
The data supporting the findings of this study are freely available in the University of Sheffield repository with the identifier https://doi.org/10.15131/shef.data.14744121.

Code availability
The custom codes used in this study are available from the corresponding author on reasonable request.

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Author contributions
P.M.W. and D.N.K. conceived and designed the experiment. T.K., P.M.W. and T.D. built the experimental apparatus and performed the experiments. P.S-J, N.C.Z., A.A., S.R. and J.B. designed and characterized the sample. A.L, I.S, L.L and A.H fabricated the sample. P.M.W. analysed the data, and wrote the manuscript and Supplementary Information with contributions from T.K. and O.K. O.K. developed the quantum theoretical description of XPM CPHASE gates. P.M.W. developed the classical theory for cavity occupancy and XPM polariton polarization rotation. All authors contributed to discussion of the data, and discussion and revision of the manuscript.

Competing interests
The authors declare no competing interests.

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