Variable potentials for thermalized light and coupled condensates

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Quantum gases in lattice potentials have been a powerful platform to simulate phenomena from solid-state physics, such as the Mott insulator transition. In contrast to ultracold atoms, photon-based platforms, such as photonic crystals, coupled waveguides or lasers, usually do not operate in thermal equilibrium. Advances towards photon simulators of solid-state equilibrium effects include polariton lattice experiments and the demonstration of a photon condensate. Here, we demonstrate a technique to create variable micropotentials for light using thermo-optic imprinting of a dye-polymer solution within an ultrahigh-finesse microcavity. We study the properties of single- and double-well potentials, and find the quality of structuring sufficient for thermalization and Bose–Einstein condensation of light. The investigation of effective photon-photon interactions along with the observed tunnel coupling between sites makes the system a promising candidate to directly populate entangled photonic many-body states. The demonstrated scalability suggests that thermo-optic imprinting provides a new approach for variable microstructuring in photonics.

Periodic potentials for light are at the core of proposals for Mott insulator physics for light, topological effects and driven-dissipative phase transitions. Exciton-polariton experiments, involving mixed states of matter and light under conditions of strong coupling, have used permanent semiconductor microstructuring, such as molecular beam epitaxy, metal depositing techniques and mirror patterning, to demonstrate double-well and periodic potentials. In the regime of weak light–matter coupling, thermalization and Bose–Einstein condensation of a photon gas have been achieved in a high-finesse microcavity containing dye molecules in liquid solution.

Here, we demonstrate a microstructuring technique that allows the generation of variable potentials for light within an optical high-finesse microcavity. The long photon lifetime enables the thermalization of photons and the demonstration of a microscopic photon condensate in a single localized site. We observe effective photon interactions as well as tunnel coupling between two microsites. The associated hybridization of eigenstates of the double-well system is monitored spectroscopically.

The scheme for thermo-optic imprinting of potentials is shown in Fig. 1a. Within a microcavity of finesse near 35,000, variations of the refractive index are induced through irradiation with a laser beam inducing heat from absorption in a 30-nm-thick silicon layer below one of the mirror surfaces. A thermosensitive polymer (poly(N-isopropylacrylamide)) undergoes a reversible phase transition to a phase with higher refractive index when heated above 305 K within a narrow temperature range of 0.2 K, is added to the dye solution between the mirrors. Local heating correspondingly increases the optical length between the mirror surfaces, which is equivalent to a local potential drop for a photon gas in the paraxial limit. This can be understood from the larger optical wavelength, corresponding to a smaller photon energy, required to locally match the mirrors’ boundary conditions.

The thermalization of the photon gas confined in the microcavity is achieved similarly as described previously. Briefly, the short mirror spacing effectively leads to a low-frequency cutoff for the photon gas. By repeated absorption and re-emission processes on the dye molecules, photons thermalize to the rovibrational temperature of the dye, which is near room temperature. As the spacing between longitudinal modes is of the order of the emission width of the dye (rhodamine in water solution), the thermalization process leaves the longitudinal mode number constant. Correspondingly, the photon gas is effectively two-dimensional, with only transverse modal quantum numbers varied.

The dye solution is pumped with a beam near 532-nm wavelength, chopped to 1-μs pulses with a 50-Hz repetition rate, to reduce the effects of pumping into dye triplet states. The pulse length is more than two orders of magnitude above the thermalization time. An example for the achieved spatial structuring of the confined photon gas in the thermo-optically imprinted potential, Fig. 1b shows the cavity emission for periodic patterns (left and middle) and a non-periodic pattern (right).

First, we investigated thermalization of the photon gas in a single microtrap. The top panel of Fig. 2a shows typical spectra of the emission of such a microsite for a low-frequency cutoff wavelength of 595 nm and different photon numbers in the cavity. The depth of the potential is h7.5 THz, corresponding to 1.21kB T at T = 305 K, where kB is the Boltzmann constant, and the observed mode spacing is Δ/2π ≈ 1.18 THz near the trap bottom, which reduces for the higher transverse modes due to deviations from a two-dimensional harmonic oscillator potential. In the thermalized case, we expect the mode spectrum to be Bose–Einstein distributed following

\[ n(i) = \frac{g(i)}{\exp\left(\frac{u_i - \mu}{k_B T}\right) - 1} \]

where \( u_i \) (with \( u_i = i\hbar\Omega \) for a harmonic potential) denotes the excitation energy of the \( i \)th trap level with respect to the low-frequency cutoff, \( \mu \) is the chemical potential and \( g(i) = 2(i + 1) \) is the...
To investigate the effect of thermo-optic interactions induced by both the pump beam and condensate photons\(^1\)\(^1\)\(^-\)\(^3\)\(^\text{,25,26}\), as understood for example from residual non-radiative decay channels of excited dye molecules, we have temporally resolved the microcavity emission frequency during a pump pulse (Fig. 2b). We observe an increasing blue-shift of the condensate frequency, corresponding to a decreasing refractive index of the dye-polymerr solution. As the temperature increases during the pump pulse, we conclude that the effective thermo-optic coefficient \(\text{dn/dT} \) is negative. The polymer in steady state has a positive thermo-optic coefficient, but due to the slow (~500 ms) timescale of its response on the 1 \(\mu\)s short timescale of a pump pulse the thermo-optic properties of the water solvent (with \(\text{dn/dT} < 0\)) dominate. Additionally, we have characterized the self-interaction of the condensate by measuring the variation of the mode diameter during one pulse (Fig. 2c). We observe an increase in diameter, and for sufficiently large pump laser spots, this variation becomes independent of details of the pump geometry, and therefore can be attributed to condensate self-interactions. Using a Gross–Pitaevskii model for the retarded thermo-optic effect, we derive a self-interaction parameter of \(\text{geff} = 2.5(8) \times 10^{-5}\) accumulated during 1 \(\mu\)s from our data (see Methods), which is even smaller than previously reported for a rhodamine–methanol medium\(^1\)\(^1\).

To study tunnelling, we employed double-well potentials with a typical spacing between microsites of 8–15 \(\mu\)m. The potentials are prepared with a well depth so shallow that only a single mode is trapped, of frequencies \(\omega_1\), \(\omega_2\) for the two microsites 1, 2, respectively. One of the sites, say site 1, is pumped, and we slightly red-shift the initial emitter frequency of this pumped site with respect to the non-pumped site 2. We find, as expected, that the condensate frequency is red-shifted upon the pumping of site 1, but due to the slow \((\sim 3\text{ ms})\) timescale of its response on the 1 \(\mu\)s timescale of a pump pulse, the condensate frequency is not significantly modulated by the pump laser spots. The observed critical photon number of 67.8(1.6) is three orders of magnitude lower than in previous work\(^\text{24}\), which is even smaller than previously reported for a photon microtrap 1\(^\text{1}\), and within uncertainties agrees with the expected value \(N_c = 67.5\) (see Methods). The observation of thermalized spectra shows that in photon microtraps loss through, for example mirror transmission, can be kept sufficiently low that within the cavity lifetime photons relax to a near equilibrium distribution. The observed condensate diameter of \(\approx 3\ \mu\m\) is close to the harmonic oscillator ground mode size, and we typically use a 15-\(\mu\)m pump beam diameter. For the future, such photon microcondensates are expected to be attractive systems to study few-particle physics effects\(^4\).

Figure 1 | Set-up and lattice realizations. a, Experimental approach to realize variable potentials for photons within a high-finesse microcavity. Optical radiation is absorbed in a thin silicon layer placed below one of the cavity mirror’s dielectric coatings. The induced heat locally increases the index of refraction of the dye-polymerr solution within the microcavity, and enhances the optical length. This results in an effective local attractive potential for the photon gas. Complex trapping potentials are realized by scanning an external ‘heating’ laser beam to imprint the desired transverse temperature modulation onto the absorbing silicon layer. The microcavity is pumped from the reverse side, and its emission can be analysed spatially, interferometrically and spectrally. AOM, acousto-optic modulator; PMT, photomultiplier tube; ICCD, intensified CCD camera; \(I_1\), \(I_2\), optical intensities; \(E_1\), \(E_2\) corresponding field amplitudes. b, Experimentally observed images of the microcavity emission for the photon gas in lattices with rectangular (left) and hexagonal (middle) geometry, respectively, and a non-periodic pattern showing the letters ‘HP’ (right).
Figure 2 | Thermalization, condensation and photon self-interactions in a single microsite. a. Top: spectrometer data analysing the microcavity emission for different photon numbers in units of the critical photon number, $N_h$. Bottom: spectrally binned data accounting for the outcoupling mirror transmission to yield the trap level populations (dots), together with theoretical expectations (lines). We observe a spectrally sharp condensate peak at the position of the cavity cutoff on top of a broad thermal cloud distributed over all bound seven trap levels. For the higher trap levels, the level spacing reduces due to deviations from harmonic trapping (see inset). BEC, Bose–Einstein condensate. b. Relative emission frequency of the dye microcavity versus time, showing a linear chirp attributed to thermal lensing induced predominantly from the pump beam. The 1-μs duration of the used pump pulses is below the thermal time constant so that no saturation of the chirp is visible (see Methods). These data were recorded for a single microcsite, but with the same pump geometry as used for the measurements shown in Figs 3 and 4. Observed condensate mode size within a pump pulse versus diameter of the pump beam. The photon number in the condensate mode is kept constant for all measurements by adjustment of the pump beam power. The broadening observed for small pump spot diameters of size comparable to the ground mode is attributed to heating directly from the pump beam. For larger diameters, the variation of the optical cloud diameter becomes independent from the pump diameter, as well understood from thermo-optic interactions due to condensate photons. The latter effect is due to heating in the course of the absorption-re-emission cycles of the dye in the presence of the finite quantum efficiency. A Gross–Pitaevskii equation model is used to describe the findings (see Methods). Inset: $I_{pump}$ pump beam intensity; $V(r)$, potential. All error bars show standard deviations.

Figure 3 | Tunnelling between two microsites. a,b. Schematic of the coupling and time dependence of the emission observed from microsite 1 (top curves in b) and site 2 (bottom curves in b) for a spacing of 13.7(2) μm (dark blue lines) and 8.6(2) μm (light blue lines), respectively. As the mode energy of the pumped site 1 increases due to thermo-optic effects, the two trap levels align in energy and photons tunnel between sites. Note that the 230-MHz bandwidth of the used photomultiplier detector is not sufficient to resolve temporal oscillations between sites. In b, the top scale gives the corresponding detuning between sites assuming a linear chirp of the mode frequency of site 1 with time, see the measurement shown in Fig. 2b. The insets show camera images recorded at corresponding times. c. Extracted tunnel coupling versus distance between microsites (dots) with standard deviation (error bars), along with a fit of an exponentially decaying curve (solid line).
to that of site 2, so that during the pump pulse the thermo-optic effect induced by the pump beam will tune the sites into resonance (Fig. 3a). Corresponding data for the time-resolved fluorescence emitted from the individual microsites is shown in Fig. 3b. As the sites are tuned into resonance, we observe tunnelling between microsites, with the number of tunnelled photons again decreasing when the sites shift out of resonance. The resonance width increases for a reduced spacing between sites, as understood from the larger tunnel coupling. The coupling strength derived from the width of the resonance (see Methods) shown in Fig. 3c follows the expected exponential scaling with the distance between microsites. At the smallest investigated distance near 8 µm, the tunnel coupling reaches a value of 16.7(1.1) GHz. The influence of the coupling on the emission frequency was explored by time-resolved spectroscopy of the emission from site 1 during the frequency chirp caused by the thermo-optic effect. Figure 4a was recorded for a tunnel coupling between sites of $J/2\pi = 1.7$ GHz, where no coupling-induced splitting of eigenstates is resolved, while the measurement shown in Fig. 4b gives data for $J/2\pi = 16.7$ GHz, where a clear splitting is visible. The emission frequencies follow well the expectations for an avoided crossing with the corresponding coupling (dashed lines), as readily obtained when modelling the double-well system with a set of coupled Schrödinger equations:

$$i\hbar \dot{\psi}_1 = \hbar \omega_1 \cdot \psi_1 + \hbar J \cdot \psi_2,$$

$$i\hbar \dot{\psi}_2 = \hbar J \cdot \psi_1 + \hbar \omega_2 \cdot \psi_2,$$



where $\psi_1,2$ denote optical wavefunctions in the corresponding sites. The eigenstates of the coupled system are $\psi_\pm = (\cos(\theta)\psi_1 + \sin(\theta)\psi_2)/\sqrt{1 + \sin^2(\theta)}$, with eigenenergies $E_\pm = \hbar (\omega_1 + \omega_2)/2 \pm \sqrt{(\hbar \Delta \omega/2)^2 + J^2}$, where $\theta = \arctan(-2J/\Delta \omega)$ denotes the mixing angle and $\Delta \omega = \omega_1 - \omega_2$. Figure 4c gives the expected emission signal $\alpha |\langle \psi_1 | \psi_1 \rangle|^2$ from site 1 for $J/2\pi = 16.7$ GHz versus the detuning. The experimental data of Fig. 4b indicates a further suppression of the emission on the weaker side of the two branches where the eigenstates mostly overlap with the unpumped site, an issue attributed to photon losses.

Figure 4b (top right) shows camera images of the emission of the double-well system for mode frequencies of site 1 below (top) and above (bottom) that of site 2. For the former case, (i), we observe a non-vanishing emission in the region between sites (in the classically ‘forbidden’ region), indicating that the system here is in the symmetric eigenstate $\psi_+$, while in the latter case, (ii), no emission is observed in that region, as expected for photons in the antisymmetric state $\psi_-$. When spatially overlapping the emission of the two microsites, the observed fringe pattern shifts by $\pi$ in phase when tuning from below to above the tunneling resonance (from case (i) to (ii)). Figure 4b (bottom right) shows the different phase factors of symmetric and antisymmetric eigenstates, respectively, of the ‘photonic molecule’, which are in good agreement with expectations.

To conclude, we have demonstrated variable potentials for light in an ultrahigh-finesse optical cavity generated using thermo-optic imprinting, a technique compatible with thermal equilibration. A microscopic photon Bose–Einstein condensate with a critical photon number of 68 was observed in a single microtrap. Effective photon interactions as well as photon tunnelling and hybridization of eigenstates in a double-well potential has been achieved.

For the future, we expect novel applications of thermo-optic patterning within low-loss resonators in other fields of photonics as well. Prospects of photon condensates in lattice potentials include the population of entangled states. Unlike in ultracold gas systems, loading and cooling in our system proceeds throughout the lattice manipulation time, and when constituting the system ground state, quantum many-body states can be populated in a thermal equilibrium process.

**Methods**

Methods and any associated references are available in the online version of the paper.

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D.D., C.K., T.D., J.S. and J.K. performed the experiments. D.D., F.V., M.W. and J.K. wrote the paper. The authors declare no competing financial interests.

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Methods
Experimental set-up and procedure. The used microcavity consists of two highly reflective plane mirrors (specified reflectivity above 99.991% in the spectral range of 520–590 nm wavelength) spaced apart by a distance of Δ = 1.98 μm. We used a plane-mirror microcavity, so that effective trapping potentials for the photon gas are solely due to thermo-optic transverse index variations. The microcavity was filled with a dye solution of the thermo-sensitive polymer poly(N-isopropylacrylamide) at concentration, ρ = 8%, and rhodamine 6G dye (ρ = 0.004%, corresponding to 10⁻⁴ mol L⁻¹) dissolved in water and Ammonyx-LO (ρ = 5%) as a surfactant. Water was used as the solvent because the polymer dissolves well in it, despite the smaller quantum efficiency of the rhodamine dye in this environment (n ≈ 80–95%) compared with that obtained when ethylene glycol-solvent was used in our earlier experiments. Above 32.4 °C, within a temperature range of ±0.2 °C, the polymer undergoes a phase transition to a gloop state with collapsed polymer chains, and the associated molecular transport increases the specified refractive index from n ≈ 1.35 to n = 1.46 (ref. 29). We have not seen evidence for a reduction of the cavity finesse caused by the dye–polymer phase transition, which we attribute to the fact that slightly above the phase transition the formed globsules for the relevant timescales remain at a relatively small size.20 The timescale of the refractive index variation within the microcavity environment was experimentally determined by monitoring the induced variation of the cavity emission frequency on local heating to be Δn ≈ 0.0005 nm. Between the glass substrate and the dielectric coating of one of the cavity mirrors a microcavity. Due to the high reflectivity of the dielectric coating, the beam near 532 nm wavelength of 5-GHz spectral resolution. We observe a nearly linear frequency chirp to higher frequencies which within the pulse time does not approach a steady state. The refractive index change due to the optical phase transition is much larger and of different sign than refractive index variations from the thermo-optic effect in water. The typical timescales are 0.1 s for a refractive index change due to the phase transition and τ = 4 ms for the usual thermo-optic effect (τ was evaluated at a few micrometres optical beam diameter). This allows the dynamic response to be dominated by the latter effect. Note that the timescale of the thermo-optic effect is dependent on the geometry of the system, and increases for larger optical beam sizes. Figure 2b shows the corresponding experimentally observed increase of the condensate frequency within the 1-μs pulse. The conclusion that the transient response is dominated by the thermo-optic effect in water is supported by the fact that within the 1-μs-long pump pulse, we observe for all pump beam diameters (that is, both in the case of the observed thermo-optic effect being dominated by heating from the pump pulse itself and the cavity photons) an increase of the ground mode diameter, as expected from Einstein condensation theory, carried out for different timescales.

The measurements shown in Fig. 2c were conducted to characterize thermo-optic effects in the dye–polymer cavity environment in more detail. For those measurements, potentials with reduced depth were used, so that only a single level was trapped (as for the data shown in Figs 3 and 4). In the experiment, the diameter of the macroscopically occupied potential well was measured alternating at the beginning and the end of the 1-μs-long pump pulse, with the gate of the ICD camera set to 50 ns. The vertical scale gives the observed relative broadening of the emitted spectral width of the modes (top panel in Fig. 2a) reaches 90 GHz for the ground mode, which is above the 30-GHz resolution of the used spectrometer, and is the result of the ground mode frequency chirp to higher frequencies which within the pulse time does not approach a steady state. The broadening is above 99.991% in the spectral range of 520–590 nm wavelength. We have not seen evidence for a reduction of the cavity finesse caused by the dye–polymer phase transition, which we attribute to the fact that slightly above the phase transition the formed globsule for the relevant timescales remain at a relatively small size.20
where $V(r, t)$ is an effective trapping potential for photons and $\mu$ is the chemical potential. Thermo-optic effects are assumed to be slow with respect to the timescale on which the photon wavefunction reaches a steady state. Correspondingly, equation (2a) has the form of the stationary Gross–Pitaevskii equation with a residual time-dependence due to the slow variation of the thermo-optical self-interaction. The interaction energy due to the refractive index change of the solution from heating through condensate photons can be written as

$$F_{\text{int}} \approx -m_{\text{ph}}(c\hbar)^2 \frac{\Delta n}{n_0}$$

where $n_0$ is the refractive index of the solution at temperature $T_0$ of the mirrors, which are regarded to act as a heat sink. Further, $\Delta n = (\text{dn/dT}) \cdot \Delta T \approx 2 \cdot (T(r, t) - T_0)$, where $\Delta n(r, t)$ and $T(r, t)$ denote the refractive index variation (with $\Delta n \ll n_0$) and temperature, respectively, at the corresponding transverse position averaged over the cavity length, and $A = (\text{dn/dT})$ is the thermo-optic coefficient of the dye solution. As we do not observe evidence for an (ultrafast) Kerr effect, we derive an accumulated effective interaction constant $g_{\text{eff}} = \frac{2(2) \times 10^{-12}}{(3) \times 10^{-16}}$, when using the above quoted value of $\tau = \tau_1 = 4 \mu$s thermal time constant. These quoted values for the dimensionless interaction constant are below that reported in earlier experiments of our group for a different experimental situation \(^{11}\), as understood mainly (i) from the use of water as a solvent in the present experiment with its comparatively low thermo-optic coefficient, (ii) the cutoff being at a relatively long wavelength resulting in a small photon absorption rate, and (iii) the here quoted value of $g_{\text{eff}}$ referring to the influence of the thermo-optic interaction during a single pump pulse.

The tunneling couplings $j$ between potential wells were determined from the spectra shown in Fig. 3b using a Bloch equation model accounting for a pump rate $\lambda$ of site 1 and a loss rate $\Gamma_1$:

$$\dot{\rho}_{11} = -\Gamma_1 \rho_{11} - 2\text{Im}(\rho_{12})$$

$$\dot{\rho}_{21} = -\Gamma_2 \rho_{21} - 2\text{Im}(\rho_{12})$$

$$\dot{\rho}_{12} = -\Delta \omega + \Gamma_2 \rho_{12} - 2\text{Im}(\rho_{12} = \rho_{21})$$

with $\rho_{12} = \rho_{21}^\dagger$, which correspond to the sites 1,2, respectively, $\rho_{12}^\dagger$ being the corresponding off-diagonal matrix element, and $\Delta \omega = \omega_2 - \omega_1$. One finds the stationary analytic solutions

$$\rho_{11}^{\text{stat}} = \frac{1}{\Delta \omega + \Gamma_1} \left( 1 - \frac{2\Gamma_2}{\Delta \omega + \Gamma_1} \right)$$

$$\rho_{12}^{\text{stat}} = \frac{1}{\Delta \omega + \Gamma_1} \left( \frac{2\Gamma_2}{\Delta \omega + \Gamma_1} \right)$$

with the saturation broadened resonance width $\Gamma_s = \sqrt{\Gamma_1^2 + \Gamma_2^2}$. From the observed value for the resonant tunneling ratio for the spectra as shown in Fig. 3b we determined the ratio $I/I_0$ of tunnel coupling and loss rate using the expected ratio $\rho_{12}(\Delta \omega = 0)/\rho_{12}(\Delta \omega = \omega_1) = 1 - \frac{1}{\Delta \omega + \Gamma_1}$ for each spectrum of a certain distance between microscopcites. This allowed us in the next step to determine the corresponding value for the tunnel coupling $J$ from the fitted width of the spectra (Fig. 3c). This readily also allowed us to determine the loss rate, and after averaging over the results of the different spectra a value $\Gamma = 11(5)$ GHz was obtained.

The spectral data shown in Fig. 4b,c was obtained by using a gated ICCD camera detector (20 ns gate time) monitoring the emission after the echelle monochromator to analyse the microcavity emission. Such measurements were repeated for different delay times after the onset of the pump pulse to allow for the analysis of the temporal variation of the microcavity emission during the frequency chirp of the detuning of microsite 1 relative to site 2 from the thermo-optic effect. The expected value for the population of dressed eigenstates of the double-well system shown in Fig. 4c was derived from projecting the eigenstates of the system onto the left well (site 1). This method gives the expected value for the relative values of the emission of the two sites in the absence of losses.

Data availability. The data that support the plots within this paper and other findings of this study are available from the corresponding authors upon reasonable request.
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