Thermally condensing photons into a coherently split state of light

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The quantum state of light plays a crucial role in a wide range of fields, from quantum information science to precision measurements. Whereas complex quantum states can be created for electrons in solid-state materials through mere cooling, optical manipulation and control builds on nonthermodynamic methods. Using an optical dye microcavity, we show that photon wave packets can be split through thermalization within a potential with two minima subject to tunnel coupling. At room temperature, photons condense into a quantum-coherent bifurcated ground state. Fringe signals upon recombination show the relative coherence between the two wells, demonstrating a working interferometer with the nonunitary thermodynamic beam splitter. Our energetically driven optical-state preparation method provides a route for exploring correlated and entangled optical many-body states.

achieving order by populating the ground state of composite systems, such as in the formation of the low-energy bonding superposition state of the H₂-molecular ion or the singlet ground state of atomic helium, is the experimental basis of many quantum phenomena, including the emergence of strongly correlated electron physics upon the refrigeration of solids (1). The observation of phenomena such as superconductivity or the fractional quantum Hall effect also depends on this approach. Cooling to the ground state is the prime example for an irreversible temporal evolution that leads to a well-defined quantum state. For light, by contrast, reversible unitary processes (such as a beam splitter) are the standard manipulation tools, with irreversibility only possible when introducing photon loss (2). For classical physics phenomena, loss can be balanced with gain, and laser-like nonequilibrium processes efficiently achieve macroscopic population of excited-state modes (3–5). Entangled few-body states of light can be created through parametric down-conversion, which requires careful postselection when aiming at more than two entangled particles (6). Advances toward the ordering of complex optical quantum systems include the observation of exciton-polariton and photon Bose-Einstein condensates (BECs) (7, 8). Although not thermalizing into the ground state of the multimode problem, recent work has demonstrated periodic potentials for both polariton and photon microcavity systems (9–12) and reported simulations of the classical XY model (13, 14). The fast time scales, in the picosecond regime for both photons and polaritons, make it impractical to temporally alternate between cooling and trap manipulation phases for optical quantum gases. Although this technique is used in the study of cold atoms in lattices, the required adiabatic mapping of the condensate state into the lattice is a main bottleneck to reach ultralow entropy states in those systems (15).

We thermalized light in a potential with two minima. Through contact to an external reservoir—that is, by means of an irreversible process—the system reaches a coherently split state. To achieve this, a double-well superimposed with a shallow harmonic trapping potential is tailored by shaping mirror surfaces in an optical dye microcavity.

In our experiment (Fig. 1A), photons were trapped in a dye-filled microcavity. Two-dimensional photon gases in such dye microcavities show a thermodynamic phase transition to a macroscopically occupied ground state, the BEC (8, 15). To engineer the desired potential, one of the two ultrahigh reflectivity cavity mirrors (cavity finesse near 100,000) has a microstructured surface profile, manufactured by use of a demetallation technique (18). In brief, the profile is created by scanning an auxiliary laser beam transversely over the mirror plane, inducing heat from absorption in a 30-nm-thick silicon layer placed below the dielectric coating (12). The heating causes a controlled local delamination of the reflective surface, which for the cavity results in a shorter distance between mirrors at the corresponding transverse position. In the paraxial limit, this induces a repulsive potential for light, which intuitively means that shorter-wavelength (higher-energetic) photons are required to match the boundary conditions of the reflecting surfaces. A spatial profile of the structured mirror profile used for this work is shown together with a cut and the calculated potential for photons within the microcavity in Fig. 1B. Near the mirror center, two indents spaced by 13 μm cause a double-well potential sufficiently shallow that only a single eigenfunction per site is trapped. Tunneling between the sites leads to a coupling of the localized wave functions, which hybridize to a symmetric \( \psi_s = \frac{1}{\sqrt{2}} (\psi_1 + \psi_2) \) and an antisymmetric wave function \( \psi_a = \frac{1}{\sqrt{2}} (\psi_1 - \psi_2) \), where \( \psi_1 \) and \( \psi_2 \) denote the wave functions localized in the wells (Fig. 1C).

The eigenenergies of the hybridized wave functions are split by \( \hbar \omega \), where \( \hbar \) is Planck’s constant divided by 2π and \( \Lambda \approx 2 \pi \cdot 30 \text{GHz} \) denotes the tunnel coupling, with the symmetric state \( \psi_s \), which is akin to the bonding state in molecular physics, being energetically lower than \( \psi_a \) (the antibonding state). To allow for Bose-Einstein condensation in two dimensions, the double-well potential is superimposed with a weak harmonic trapping potential (18).

Thermalization of photons was achieved by following methods described previously (8, 19). The two cavity mirrors, because of their small spacing in the micrometer regime, impose an upper limit of the optical wavelength that fits inside the resonator, corresponding to a restriction of energies to a minimum cutoff of \( \hbar \omega = 2.1 \text{eV} \), which we identify here with the eigenenergy of the bonding state \( \psi_s \) of the double well. Moreover, the optical dispersion of cavity photons in the paraxial limit becomes quadratic, the same as it would for a massive particle. Given that interparticle interactions are weak, (number-conserving) thermalization of photons was achieved by means of repeated absorption re-emission processes on the dye molecules, which occurred to the vibrational temperature of the dye, which is at room temperature. Because of the frequent collisions of solvent molecules with the dye (18, 19), photons emitted by the dye molecules were thermally equilibrated in an irreversible process. Because of the large mode spacing in the wavelength-spaced cavity, order of the emission width of the dye, thermalization leaves the longitudinal mode unchanged, whereas the remaining transverse degrees of freedom make the photon gas two-dimensional. It has been shown (19) that the system is formally equivalent to a two-dimensional system of trapped massive bosons with effective mass \( m_{ph} = \hbar \omega_0 n^2/c^2 \), where \( n \) is the refractive index of the dye solution and \( c \) is the vacuum speed of light. To inject photons and compensate for losses, the dye is weakly pumped with a laser beam. Despite pumping and losses, the photon gas well follows an equilibrium Bose-Einstein distribution

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

because photons thermalize faster than they are lost through, for example, mirror losses (20, 21).

Here, \( u_i \) is the excitation energy above the cut-off, \( g_i \) is the mode degeneracy, \( \mu \) is the
Fig. 1. Experimental environment. (A) Photons are trapped in a microcavity with a mirror spacing corresponding to an optical path of 5.5 wavelengths, where one mirror is laterally microstructured. The photons thermalize through radiative contact to the dye molecules (bottom schematics). $D_0 = q \frac{\lambda}{2}$ describes the mirror spacing, where $q$ is the longitudinal mode number and $\lambda$ is the optical wavelength. (B) Height profile of the microstructured mirror surface (main plot) and profile of the corresponding expected trapping potential for cavity photons (right), realizing a double-well structure in the center superimposed with a harmonic trapping potential. (C) Schematic energy level structure, with the symmetric eigenstate of the double well as the lowest energetic eigenstate.

Fig. 2. Spectrally resolved measurements. (A) Measured wavelength versus transverse position along the axis of the double well both below (top, $N/N_{c,\text{exp}} = 0.94$) and above (middle, $N/N_{c,\text{exp}} = 1.05$) the threshold to Bose-Einstein condensation (BEC). $N$, measured total photon number; $N_{c,\text{exp}}$, measured critical photon number. The high-resolution spectra cover the range of the first nine lowest-energetic (highest-wavelength) modes. From right to left, the first two modes are the symmetric and antisymmetric modes of the double well, followed by modes not confined in the wells, which for the higher-order modes are well described by harmonic oscillator modes with a trap frequency of $\omega = 2\pi \times 63(2)$ GHz. The enhanced emission of the symmetric superposition observed in the latter image is attributed to Bose-Einstein condensation. a.u., arbitrary units. (B) Potential Range. (Bottom) Spectra for different photon numbers. (B) Broadband, low-resolution spectra for different photon numbers (dots), along with theory for the dye microcavity temperature $T = 300$ K. The observed width of the BEC peak is dominated by spectrometer resolution. The spectral position of the rim of the potential well is indicated with the dashed line. For all measurements, the critical photon number is $N_{c,\text{exp}} = 8000$. 
chemical potential, $k_B$ is the Boltzmann constant, and $T$ is the temperature. For the lowest two levels of our system, the bonding and antibonding states of the double well, we have $u_0 = 0$ and $u_a = \hbar\Delta$, with $\Delta = 2$ in both cases because of polarization degeneracy [details of our theoretical modeling are provided in (18)].

In the experimental data analyzing the microcavity emission, Fig. 2A, top and middle, gives color-coded high-resolution spectra spatially resolved along the direction of the double-well axis. Shown in Fig. 2A, top, are data recorded in the thermal regime with a photon number far below the Bose-Einstein condensation threshold. On the long-wavelength (low photon energy) side, we observed emission of the antisymmetric (slightly below 586 nm) and symmetric (slightly above 586 nm) double-well modes. In the former case, the emission in the center vanishes, indicating photons in the antisymmetric state, whereas the finite intensity in the center for the latter case corresponds to a population of the symmetric wave function. To verify the thermal distribution of modes, we recorded broadband spectra for different total photon numbers; see Fig. 2B, along with theory curves (Fig. 2B, solid lines), for a Bose-Einstein distribution (Eq. 1). The bottom curves in Fig. 2B correspond to the thermal regime, whereas at high photon numbers, we observed a peak at the position of the cavity low-frequency cutoff near 586 nm, which we attribute to photon Bose-Einstein condensation. The experimental data are in good agreement with the equilibrium theory for 300 K down to a wavelength near 582 nm, at which we approach the finite rim of our imprinted potential (Fig. 1B) at $\sim 0.54 \cdot k_B T$ above the cutoff energy, owing to present limits of our delamination technique. The thermal cloud saturates for photon numbers above the BEC threshold, verifying a central prediction of BEC theory.

Data above the condensation threshold are shown in Fig. 2A, middle, with the individual cavity mode energies spectrally resolved (Fig. 2A, bottom, green data points). The visible strongly enhanced emission at the position of the cavity low-frequency cutoff is evidence for Bose-Einstein condensation of photons into the symmetric superposition $\psi_s$ of the two microsites’ eigenfunctions, where the condensate contains approximately 1200 photons (Fig. 2A, bottom, red data points). The energetic spacing to the first excited state ($\psi_a$) is $\hbar\Delta \approx k_B \cdot 1.4$ K, which is roughly a factor of 200 below the thermal energy of the room-temperature system. This comparison, in addition to the good agreement of our data with BEC theory, makes it clear that the observed population enhancement was a consequence of quantum, rather than classical, statistics.

The full spatial profile of the cavity emission is shown in Fig. 3A both below (Fig. 3A, left) and above (Fig. 3A, right) the critical photon number. Corresponding cuts along the direction of the double-well are shown in Fig. 3B for...
both the data below (Fig. 3B, blue line) and above (Fig. 3B, black line) criticality, where in the latter case the split spatial profile of the condensate is clearly visible. To determine the relative phase of the emission originating from the individual microsites, we directed the cavity output through an optical interferometer. Camera images are shown in Fig. 4A of the resulting signal recorded in the thermal (Fig. 4A, left) and the condensed phase (Fig. 4A, right), respectively; the marked central areas in Fig. 4A, left and right, correspond to the region where the emission of the individual microsites spatially overlap. In this region, we observed a stable interference signal for the condensed phase (Fig. 4B), even though the shown data are the average of many subsequent experimental realizations (Fig. 4) (18). This verifies the fixed phase relation between the microsites, as expected for a delocalized, macroscopically occupied superposition state. In other words, the observation of a fringe pattern sets an upper limit on the indistinguishability (“which-path information”) left in the individual microsites upon thermalization. This can be understood from the tunneling time $\pi/\Delta \approx 17$ ps being below the $\approx 100$ ps dye reabsorption time during the absorption and emission cycles for the used experimental parameters (21).

Our observation of Bose-Einstein condensation of photons into the bonding low-energy superposition state of a double-well potential demonstrates the irreversible but coherent population of split states of light, in which the bifurcated single-photon state is massively occupied because of Bose-Einstein statistics.

Thermalization of light to cold reservoirs in tailored trapping geometries provides a possible route for the direct preparation of more complex quantum states. Strong photon interactions are expected from using second-order nonlinear materials in a doubly resonant cavity setup, yielding a controllable effective Kerr interaction (22). When in a periodic lattice potential quantum many-body states then become the ground state; they then can be selectively populated through thermalization (18, 23–26), which is not yet achievable in present atomic physics experiments (15). For analysis, optical many-body state tomography can be performed with correlation measurements (6). Other perspectives of our work include the exploration of correlated quantum states in both double-well and periodic potential lattice systems coupled by means of effective particle exchange to the photoexcitable dye molecules (27, 28). Last, applications of the described delamination-based adaptive optics method can range from gain-dissipative simulations of the classical XY model in partial equilibrium condensate arrays (13, 14) to optical phase holography (3).

REFERENCES AND NOTES

18. Supplementary text is provided as supplementary materials.

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Data and materials availability: Data shown in the figures are available on the Zenodo public database (29).

SUPPLEMENTARY MATERIALS

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Fig. S1

References (30–40)

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Irreversible splitting of light
Prisms and dielectric beam splitters tend to be unitary and reversible optical elements, with the quantum properties of the photons largely irrelevant. Kurtscheid et al. introduce a method of irreversibly, but coherently, populating a split state with photons by thermalizing the photons into a low-energy ground state by repeated absorption-emission interaction with a fluorescent dye within a double-dimple optical cavity. Generation of such a coherent split state could be used as a precursor step to the quasi-continuous creation of many-body entangled states of light, which could be useful in applications in quantum communication, computing, and simulation. Science, this issue p. 894