Unsupervised Learning of the Fuzzy Phases of Small Photonic Condensates

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Phase transitions, while ubiquitous in nature, are formally defined only in the thermodynamic limit. While criticality is well approximated in large systems, large relative fluctuations in systems made up of only a few particles strongly hinder our ability to identify and characterize different phases of matter. Here, we demonstrate that unsupervised learning and fuzzy logic permit the detection of the otherwise inaccessible and subtle phase structure of small physical systems, in a data-driven and model-free approach. We thus introduce the concept of fuzzy phases and, in particular, construct the fuzzy phase diagram of a photonic condensate made up of only a few photons. The notion of thermodynamic phases and phase transitions is therefore generalized into the realm of finite, and particularly small, physical systems.

Phase transitions are extraordinary manifestations of collective behaviour that mark abrupt changes in the properties of many-particle systems. The associated discontinuities in the appropriate thermodynamic quantities can be mathematically described by the so-called Yang-Lee singularities, which only emerge in the limit of infinite degrees of freedom. Intuitively, extensive quantities $Q$, like energy or particle number, have fluctuations of the order $\sqrt{Q}$, with $\sqrt{Q}/Q$ vanishing in the thermodynamic limit $Q \rightarrow \infty$, giving rise to sharp transitions. Up until recent years, and in spite of the abstractness of the thermodynamic limit, experimental systems of interest were often large enough such that quasi-critical behavior was observed. This usually comes in the form of diverging susceptibilities or heat capacities, allowing the unequivocal detection of a phase transition.

The identification of phase transitions may still be hindered by a high-dimensional configuration space, or the existence of nontrivial order parameters, like in topological or many-body localized states. Machine learning techniques, such as neural networks, have been shown to successfully detect and label such complex phases of matter, mostly due to their ability to retrieve the often few significant features in otherwise large sets of data. These frameworks, however, require prior knowledge of the phase structure of the system’s Hamiltonian, falling into the domain of supervised learning, with few experimental results yet reported. A few examples include the training of a neural network far from the critical region and the posterior characterization of the Mott insulator-superfluid transition, or the usage of an artificially synthesized dataset, carefully designed to reflect the expected symmetries of a nematic phase in electronic quantum matter.

Photonic condensates are powerful platforms for exploring the fundamental physics of phase transitions and critical phenomena. Equilibrium Bose-Einstein condensation of light has been achieved in diverse platforms, including semiconductor microcavities, dye-filled microcavities, plasmonic lattices, or fibre cavities. Dye-filled microcavities are particularly interesting, as driving, loss and thermalization rates can be independently controlled to give access to rich non-equilibrium behavior. The further ability to precisely engineer the trapping potential results in an impressive degree of control over these systems.

Here, we explore the phases of a small photonic condensate in a dye-filled microcavity – see Materials and Methods section for more details.
The trapping potential is engineered to combine a small cavity volume with a large mode spacing, as shown in the Methods section for further details on the experiment. Our multi-mode microcavity operates in the thermodynamic limit, i.e. the thermodynamic limit $\beta_{\text{Th}}$. Criticality is thus recovered in the large cavity limit, i.e. the thermodynamic limit $\beta_{\text{Th}}$. The parameter $\beta_{\text{Th}}$ is defined as the fraction of spontaneous emission into the $m$th cavity mode, generalizing the standard $\beta$ parameter introduced in the context of single-mode microlasers into the realm of multi-mode systems. In the absence of mode-mode coupling and photon re-absorption, the lasing, or condensation, transition occurs at a photon number that scales as $\beta_{\text{Th}}^{-1}$, while the corresponding threshold width is of the order $\sqrt{\beta_{\text{Th}}}$. Criticality is thus recovered in the large cavity limit, i.e. the thermodynamic limit $\beta_{\text{Th}}^{-1} \to \infty$. More details can be found in the Methods section. Our multi-mode microcavity operates in a mesoscopic regime characterized by the existence of collective behavior despite the small system size. While smooth crossovers between qualitatively different states will be identified, the mesoscopic regime and the inherent large relative fluctuations of order $\sqrt{Q}/Q$ strongly contrast with the idea of phase transitions being only defined in the thermodynamic limit.

The photonic modes inside the microcavity are those of a two-dimensional harmonic oscillator, each with a degeneracy proportional to the mode number $m$. There is a single $m=0$ ground-state located at approximately 540 THz or, equivalently, 560 nm. Excited states are separated by roughly 2.1 THz, which is only slightly smaller than the thermal energy scales, with only a few low-energy modes becoming thermally accessible. More details can be found in the Materials and Methods section. Photon thermalization results from multi-mode emission and absorption events from and by the dye molecules. These occur at rates $E_m$ and $A_m$, respectively, which are related by the Keldysh-Stepanov relation $A_m = E_m e^{\gamma m/k_B T}$. Here, $\gamma = \omega_{ZPL} - \omega_m$, with $\omega_{ZPL}$ denoting the zero-phonon line of the dye molecules.

We work on the Stokes side of the molecular transition, where $E_m > A_m$. Constant pumping of dye excitations is required to maintain steady-state operation due to the finite mirror transmission, which is quantified by the cavity loss rate $\kappa$. Since the finite cavity lifetime limits the thermalization process, we define the thermalization coefficient $\gamma = A_0/\kappa$ as the average number of absorption events per cavity lifetime, with $A_0$ the absorption rate at the cavity cutoff. Regimes of good thermal contact with the molecular reservoir imply fast thermalization, i.e. $\gamma \gg 1$. 

FIG. 2. Occupation numbers. A-C, Absolute and relative occupations numbers of the first four photonic modes, for different thermalization regimes. The latter is experimentally tuned by sweeping the cavity cutoff wavelength $\lambda_0$. Each set of occupation numbers defines a configuration. The dashed vertical lines mark transitions between different representative phases, which are labeled as: Th: thermal phase; BEC: Bose-Einstein condensate; L1: condensed first excited-state; L2: condensed second-excited state. D-F, Results from the non-equilibrium model of photonic condensation.
Besides the driven-dissipative character described above, additional processes occurring in the microcavity contribute to the emergence of complex non-equilibrium behavior, as schematically depicted in panel (A) of Fig. 1. The heterogeneity among the different mode functions gives rise to a spatially-dependent competition for the finite molecular excitations. The result is a form of incoherent mode-mode coupling mediated by dye molecules, an effect which becomes more noticeable at higher pump powers and responsible for the breaking of thermal equilibrium.

By tuning the thermalization coefficient and the pump power, we can thus bring our photonic system into distinct equilibrium and non-equilibrium regimes, as shown in Fig. 2. Regions of good thermal contact are characterized by the existence of a wide thermal regime followed by a smooth transition into a state where most photons occupy the ground-state alone, consistent with a Bose-Einstein condensate. On the contrary, under weaker thermal contact excited states are shown to become highly populated as the pump power is increased, indicating a breakdown of thermal equilibrium. A non-equilibrium model derived from a full quantum description accurately describes these observations, particularly in the regimes of strong thermalization. Further details on the model can be found in the Materials and Methods section. For weaker thermalization, the mode condensation dynamics are highly dependent on imperfections in the shape and alignment of the pump beam, which is at the origin of the slight deviations between theory and experiment depicted in Fig. 2.

Despite the qualitative observations above, systematically inferring the existence or not of different phases remains challenging. Certainly, any form of critical behavior is absent. This is neither a particularity of the system at study or a limitation of the measurement apparatus, but rather a fundamental statistical consequence of the small particle number and the complete breakdown of the thermodynamic limit. This begs the question of whether the traditional concept of phase transitions is of any use this far from the thermodynamic limit. We tackle this question here by turning to a machine-augmented approach and, as a result, introduce the concept of fuzzy phases. Importantly, the common supervised learning techniques require the prior knowledge of the Hamiltonian phase structure, used to train models capable of

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**FIG. 3. Fuzzy phase diagram.**

- **A.** Total photon number as a function of pump power and thermalization coefficient. Different representative phases are shown by different colors, with a transparency level proportional to the respective membership entropy. The resulting standard deviation is depicted by the dashed grey area. This ensures the stability of this procedure.
- **B.** Representative phase diagram. Different representative phases. The dashed vertical lines indicate the traces depicted in Fig. 2.
- **C.** Estimation of the number of phases by minimizing the average membership entropy, whose distribution is approximated by bootstrapping the feature space. The resulting standard deviation is depicted by the dashed grey area. This ensures the stability of this procedure.
- **D.** Membership entropy. The fuzzy phase diagram is constructed from (D) and (E) by assigning a level of transparency proportional to the membership entropy, as shown in (F), with regions of phase ambiguity becoming white, these separating different representative phases.

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FIG. 4. Fuzzy and sharp photonic condensates. B, Fuzzy phase diagram obtained from the non-equilibrium model of photonic condensation, with parameters that match the experiment. As described earlier, the model slightly deviates from the experiment at weak thermalization conditions, where it predicts the condensation of the third excited cavity mode, the L3 phase. At intermediate thermalization rates, we also infer the presence of a multi-mode (MM) phase, characterized by the condensation of both the ground and the first excited state. This is, however, a relatively narrow and fuzzy phase which is not resolved in the experiment. The theoretical model is also used to generate configurations for both smaller (A) and larger (C) photonic condensates, quantified by the fraction of spontaneous emission into the ground-state. The thermodynamic limit is approached from left to right.

inferring the phase of unlabeled configurations.\cite{11,14} By contrast, here we start completely devoid of knowledge about our photonic system, thus necessarily falling into the domain of unsupervised learning.

The task now turns into inferring structure from measured data alone. We consider the relative occupation numbers of the ten lowest-energy photonic modes. A configuration becomes a point in this bounded ten-dimensional feature space and different configurations are spanned by changing the pump power and thermalization coefficient, these last two defining the parameter space. The search for structure proceeds with the clustering of nearby points in feature space, under a similarity metric, as schematically depicted in panel (B) of Fig. 3. Each cluster contains similar configurations which are maximally distinguishable from those of the remaining clusters, being thus identifiable with a particular phase. We point out that it is important to uniformly sample the parameter space, such that all inferred structure becomes uniquely linked to the intrinsic phase properties of the system at study. The smooth transitions between phases suggests the use of fuzzy logic. Formally, for each configuration, i.e., each point \( x \) in the feature space, we wish to find the probabilities \( p_x(i) \), with \( i = 1, 2, ..., k \) and \( k \) the estimated number of phases, such that \( p_x(i) \) is the Bayesian probability of membership of configuration \( x \) to the \( i \)th phase, thus gauging a level of membership with phases. This can be achieved by the \textit{fc-means} algorithm.\cite{13,15} This unsupervised learning approach will allow us to recover the subtle phase structure of our photonic condensate in a data-driven and model-free approach.

We thus introduce the concept of fuzzy phases, which follows immediately from the fuzzy logic formalism described above. In particular, the fuzzy phase congruent with a given configuration \( x \) is fully determined by the set of membership probabilities \( p_x(i) \). Here, the fact that physical systems, in particular those with few particles, often exhibit properties simultaneously consistent with multiple phases, becomes inherent to the whole formalism, thus generalizing the notion of thermodynamic phases and phase transitions. Furthermore, the full information contained in the set of probabilities \( p_x(i) \) can be succinctly summarized by the following two quantities. The first is the representative phase which, for a fixed configuration \( x \), is taken as the phase \( i \) with the highest membership probability \( p_x(i) \), if it exists. The second is the membership entropy, which we define as

\[
S_x ([p_x(i)]) = - \frac{1}{\log(k)} \sum_i p_x(i) \log(p_x(i)),
\]

defining the ambiguity in associating configurations with phases. It is normalized such that a maximally fuzzy configuration, or maximally fuzzy phase, has unit entropy, corresponding to \( p_x(i) = 1/k \). On the other hand, definite phases are recovered in the limit \( S_x = 0 \), corresponding to \( p_x(j) = 1 \) and \( p_x(i \neq j) = 0 \).

The learned phase structure of our small photonic system is depicted in Fig. 4. Four different photonic phases are estimated by minimizing the average membership entropy, depicted in panel (C). We begin by constructing the representative phase diagram, shown in panel (D). By inspection of the bosonic occupation numbers in Fig. 2, the different phases can be associated with: a thermal phase (Th); a Bose-Einstein condensate (BEC); and the condensation of the first (L1) and second (L2) excited states. Note that, while instructive, the information contained in the representative phase diagram alone
is fundamentally incomplete. The membership entropy further complements the picture and, together with the representative phase diagram, allows us to construct the fuzzy phase diagram, depicted panel (F), which directly reflects the fundamental absence of critical behavior inherent to the few-particles regime. Here, instead, different representative phases are separated by broad regions of large membership entropy or, equivalently, large phase ambiguity.

The new concept of fuzzy phases can be further explored by considering the non-equilibrium theoretical model of photonic condensation, as depicted in Fig. 1. Here, we manipulate the system size by changing the parameter $\beta_m$. We begin by simulating a smaller system than that of the experiment, $\beta_m > \beta_{m}^{exp}$. Here, the membership entropy (fuzziness) increases across the entire phase diagram. As a result, the optimal classification essentially retains the existence of only the thermal and the Bose-Einstein condensed phase. Previously inferred phases in larger systems are now blurred together with the thermal phase, as it no longer becomes relevant to consider them as independent phases. This is not arbitrarily imposed but rather optimally inferred by the fuzzy clustering procedure acting on observational data alone.

On the other hand, for a larger system, $\beta_m < \beta_{m}^{exp}$, the membership entropy across the phase diagram becomes smaller and mostly concentrated in the increasingly narrower regions between representative phases. In the limit of infinite number of particles (thermodynamic limit), the membership entropy vanishes everywhere, with representative phases becoming definite phases, except in the infinitesimally narrow regions marking pure phase transitions, thus recovering criticality. Thermodynamic phases and phase transitions are then recovered, and can both be thought of as limiting situations of a more general framework. Fuzzy phases then unify the concept of thermodynamic phases and phase transitions into a single entity, at the same time as generalizing them to systems where criticality is fundamentally absent.

The increasing ability to control and prepare small atomic \cite{28,30,37,38} and photonic \cite{29,30,39,40} systems has originated a great deal of scientific interest in few-particle physics. Recent progress in these areas demands a redefinition of the fundamental notion of phases and phase transitions. We have introduced a new paradigm, based on the machine-augmented ability to distinguish between sets of possibly similar configurations. Here, criteria for defining phases and phase transitions are neither phenomenologically or axiomatically imposed, but rather optimally inferred from observational data alone, requiring no prior assumptions or knowledge about the system. This paradigm thus solves the ill-definition of phases and phase transitions in finite systems by introducing the concept of fuzzy phases and the resulting membership entropy measure. While the clustering approach is applicable for systems of any size, the fuzzy logic approach becomes particularly relevant in small system. Importantly, the fuzzy character does not reflect a state of incomplete knowledge but rather a fundamental statistical implication of the small particle number. Our results also demonstrate the strong synergies between the powerful frameworks of statistical physics and thermodynamics on one side, and data science and machine learning on the other side. We anticipate immediate applications of the fuzzy phases concept in the investigation of how collective effects emerge from a bottom-up approach, as the system’s size is gradually increased \cite{43} or in the investigation of how magnetic phases change in the few-particle limit \cite{44}. A distinct, yet exciting possibility, would be the exploitation of the fuzzy phases concept in the context of liquid phase condensation inside biological cells, where the formation of membrane-less coherent structures seems to depend on smoothly varying concentration thresholds, suggesting the presence of significant finite-size effects. \cite{43,44}

Materials and Methods

Experimental details

The experiment consists of an open microcavity composed of one large planar and one microfabricated spherical mirror. We typically work at the fixed tenth longitudinal mode, corresponding to an effective cavity length of approximately 1 $\mu$m. This ensures a large free spectral range, thus freezing out all longitudinal dynamics. The intra-cavity region is filled with a solution of a highly-fluorescent dye (Rhodamine-6G) in ethylene glycol. The dye is incoherently pumped with a green c.w. laser at 532 nm. The pump is transmitted through the planar mirror at approximately 52 degrees and its focal region is aligned with the centre of the spherical mirror. To avoid populating the molecular excited triplet state, the pump pulse is chopped with an acousto-optic modulator. The pulse duration is kept at 350 ns and the repetition rate set to maintain the average power below 0.1 mW, which limits photo-bleaching and permanent dye damage. The long pulse duration allows most of the acquisition to occur at steady-state operation, which is attained at the few nanosecond timescale.

The cavity effectively separates the longitudinal (one-dimensional) and transverse (two-dimensional) photon dynamics. While the former is essentially frozen, the latter is that of a massive boson in a harmonic potential of frequency $\Omega = c/n\sqrt{L_0R_c}$. Here, $c$ is the speed of light, $n$ the refractive index of the dye solution and $L_0$ the effective cavity length. The tight radius of curvature of the mirror, $R_c = 250 \mu$m, translates into a large mode spacing of approximately $\Omega = 2.1$ THz, or about 2.2 nm in the optical region. As such, given the thermal range at room temperature of approximately $k_BT/h \approx 6.3$ THz, only a few low-lying photonic modes become thermally accessible. Together with the small cavity volume and the high fraction of spontaneous emission into cavity modes, several qualitatively distinct emission regimes are spanned.
even at small photon numbers.

The tightly-sculpted spherical mirrors are fabricated using focused-ion-beam milling (FIB). Further details of the fabrication can be found elsewhere. The energy, or wavelength, of the cavity ground-state is set by adjusting the cavity length with a piezoelectric actuator. The light leaking from the planar mirror side of the cavity is directed onto a calibrated spectrometer, which has a resolution of approximately 0.3 mm (or 0.3 THz). Together with the knowledge of the mirror transmission we can spectrally resolve the ensemble-averaged occupation numbers of the full bosonic field.

Non-equilibrium model of photonic condensation

The dynamics of photons in a dye-filled microcavity can be accurately described in terms of a full non-equilibrium quantum model which takes into account the spatial degrees of freedom of the molecular reservoir. It captures several processes associated with the resulting mode-mode competition, including decondensation phase transitions and non-critical slowing down. At the mean-field level, the model is reduced to a set of rate equations for the mode occupation numbers \( n_m \), namely

\[
\dot{n}_m = -\kappa n_m + \sum_j g_{mj} E_m (n_m + 1) f_j N_j - \sum_j g_{mj} A_m n_m (1 - f_j) N_j. \tag{2}
\]

Here, \( m \) labels the different cavity modes and \( \kappa \) denotes the cavity loss rate. The emission and absorption rates associated with mode \( m \) are denoted by \( E_m \) and \( A_m \), respectively. In order to account for the spatial degrees of freedom, the molecular reservoir can be divided into a finite set of spatial bins, spanned by the index \( j \), and each containing a total of \( N_j \) molecules. In the same way, \( f_j \) denotes the fraction of excited molecules in the bin. The coupling term \( g_{mj} \) is defined as

\[
g_{mj} = \frac{|\Psi_m(r_j)|^2}{\sum_i |\Psi_m(r_i)|^2}. \tag{3}
\]

It quantifies the coupling strength between mode \( m \) and dye molecules in bin \( j \), centered at the transverse cavity plane coordinates \( r_j = (x_j, y_j) \). Depending on the specific form of the mode functions \( \Psi_m(r_j) \), the same dye molecules can couple heterogeneously to different cavity modes, which is at the origin of the coupling processes described in this work. The molecular excitation fraction is determined by

\[
\dot{f}_j = -\Gamma_{ij} f_j + \Gamma_{j}(1 - f_j), \tag{4}
\]

with \( \Gamma_{ij} \) and \( \Gamma_{j} \) the spatially dependent total rates of emission and absorption, given by

\[
\Gamma_{ij} = \Gamma_{loss,j} + \sum_m g_{mj} E_m (n_m + 1), \quad \text{and} \quad \Gamma_{j} = \Gamma_{pump}(r_j) + \sum_m g_{mj} A_m n_m. \tag{5}
\]

Here, \( \Gamma_{loss} \) is the rate of molecular decay into non-cavity modes and \( \Gamma_{pump}(r_j) \) is the spatially-dependent incoherent pump rate.

In the standard model of single-mode microlasers a \( \beta \) parameter is defined as the fraction of spontaneous emission into the cavity. We can generalize this concept into the realm of multi-mode cavities by defining the \( \beta_m \) parameter as

\[
\beta_m = \sum_j g_{mj} \left( \frac{g_{mj} E_m}{\Gamma_{loss,j} + \sum_m g_{mj} E_m} \right), \tag{7}
\]

which takes into consideration the spatially resolved character of the molecular reservoir quantified by the coupling terms \( g_{mj} \).

The computational cost of integrating Eqs. (2) and (4) can be greatly reduced by approximating the full spatial resolution of the model with a hierarchy of collective molecular modes \( \mathcal{F}_m \). Sufficiently accurate results can be obtained by truncating the hierarchy after a few orders. In the collective mode picture, Eq. (2) is rewritten as

\[
\dot{n}_m = -\kappa n_m + E_m (n_m + 1) \mathcal{F}_m - A_m n_m (N_m - \mathcal{F}_m), \tag{8}
\]

with \( \mathcal{F}_m = \sum_j g_{mj} f_j N_j \) and \( N_m = \sum_j g_{mj} N_j \). The collective molecular excitation mode is now determined by

\[
\dot{\mathcal{F}}_m = \mathcal{L}(n_0, ..., n_m, \mathcal{F}_0, ..., \mathcal{F}_m, \mathcal{F}_0, ..., \mathcal{F}_k), \tag{9}
\]

where \( \mathcal{L} \) is an algebraic function that is numerically evaluated and \( \mathcal{F}_k \) are the higher order collective excitation modes, defined in a optimally rotated spatial basis. The idea is to infer an optimal basis where the collective excitation modes are maximally decoupled, with coupling being hierarchically approximated by higher order of \( \mathcal{F}_k \). More details on the numerical implementation can be found elsewhere. The theoretical model is also used to calibrate the absolute mode occupation number, which we have previously shown to be accurate.

The full non-equilibrium model described above can, in one limit, be simplified into a structure equivalent to the standard microlaser model. We begin by assuming that the molecular reservoirs do not affect each other i.e., \( g_{mj} = \delta_{mj} \). Although this procedure would be more relevant in the rotated basis, where the reservoirs are already maximally decoupled, we perform this approximation at the level of the original Eqs. (2) and (4). In this case, Eq. (4) now reads

\[
\dot{f}_m = -[\Gamma_{loss} + E_m (n_m + 1)] f_m + [\Gamma_{pump} + A_m n_m](1 - f_m). \tag{10}
\]
In the absence of any significant photon absorption, the steady-state solution to Eqs. (8) and (10) is given by

\[ n_m = \frac{p_m - \beta_m^{-1} + \sqrt{-4p_m(\beta_m^{-1} - 1) + (p_m + \beta_m^{-1})^2}}{2}, \]

where \( p_m = \Gamma_{\text{pump}}N_m/\kappa \) is the normalized pump rate. Here, we naturally recover the \( \beta_m \) parameter as \( \beta_m = E_m/(E_m + \Gamma_{\text{loss}}) \), which could be directly obtained from Eq. (7) and the assumption of independent reservoirs \( g_{mj} = \delta_{mj} \). The dynamics of the multi-mode cavity is then reduced to that of a collection of independent photonic modes. The microlaser limit corresponds to \( \beta_m \rightarrow 1 \), when most of the molecular emission goes into the cavity mode. This is accompanied by a complete lost of criticality as the system transitions from spontaneous into stimulated emission, as it has been described in the context of thresholdless lasers.[2252]

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Author contributions

J.D.R. and R.A.N. conceived the experiment; J.D.R. carried out the experiments and analyzed the data, with assistance from B.T.W., R.A.N. and R.F.O; H.S.D., J.D.R. and B.T.W. worked out the theory and ran the simulations, with assistance from F.M.; B.T.W. and J.M.S. fabricated the mirrors; J.D.R. wrote the manuscript with input from all authors.
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