Exciton–polariton condensates

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Recently a new type of system exhibiting spontaneous coherence has emerged—the exciton–polariton condensate. Exciton–polaritons (or polaritons for short) are bosonic quasiparticles that exist inside semiconductor microcavities, consisting of a superposition of an exciton and a cavity photon. Above a threshold density the polaritons macroscopically occupy the same quantum state, forming a condensate. The polaritons have a lifetime that is typically comparable to or shorter than thermalization times, giving them an inherently non-equilibrium nature. Nevertheless, they exhibit many of the features that would be expected of equilibrium Bose–Einstein condensates (BECs). The non-equilibrium nature of the system raises fundamental questions as to what it means for a system to be a BEC, and introduces new physics beyond that seen in other macroscopically coherent systems. In this review we focus on several physical phenomena exhibited by exciton–polariton condensates. In particular, we examine topics such as the difference between a polariton BEC, a polariton laser and a photon laser, as well as physical phenomena such as superfluidity, vortex formation, and Berezinskii–Kosterlitz–Thouless and Bardeen–Cooper–Schrieffer physics. We also discuss the physics and applications of engineered polariton structures.

Spontaneous coherence is a phenomenon that has fascinated physicists from a wide range of fields, ranging from condensed matter physics, atomic physics and quantum optics, to high-energy physics. Lasing is perhaps the most ubiquitous phenomenon giving rise to macroscopic coherence, in this case formed by stimulated emission of photons¹. Bose–Einstein condensation (BEC) is another example of collective coherence of many particles, such that above a critical density (or equivalently below a critical temperature), the particles spontaneously occupy the ground state². Superfluid³ He is the earliest realization of BEC, in the presence of strong interactions⁴. Superconductivity, viewed as a condensation of Cooper pairs, allows a charged version of BEC yielding resistanceless (superfluid) flow⁵. The aspect that is common to these phenomena is that a large number of particles initially possessing no phase relation all become coherent once a system parameter, such as temperature or density, crosses a threshold. In this review, we examine a new system that undergoes spontaneous coherence: the exciton–polariton condensate. The recent observation of exciton–polariton condensation⁶–⁷ adds another particle to the list for which BEC has been observed—cold atoms⁸–⁹, magnons¹⁰,¹¹, and more recently photons¹². We shall see that rather than being simply another type of particle that undergoes BEC, it possesses characteristics that incorporate new physics owing to its intrinsically non-equilibrium nature.

One of the distinctive features of exciton–polaritons (or simply polaritons for short) is their exceedingly light effective mass, typically of the order of 10⁻⁴ times the bare electron mass. For an ideal (non-interacting and at thermal equilibrium) bosonic gas in three dimensions, the critical temperature for BEC occurs when \( n^c = \frac{\lambda_d^3}{\pi^2 n} = 2.62 \), where \( n \) is the density of the bosons and \( \lambda_d = \sqrt{\frac{\pi T}{m k_B T}} \) is the de Broglie wavelength (\( m \) is the mass of the bosons, \( k_B \) is Boltzmann’s constant and \( T \) is the temperature). This criterion can intuitively be thought of as being when the density of the bosons is high enough such that their wavefunctions overlap. The mass dependence of \( \lambda_d \) means that it is easier to produce Bose–Einstein condensates (BECs) for light mass particles, which is one of the great advantages of using polaritons. For most experiments so far, exciton–polariton condensates are produced at cryogenic temperatures in the vicinity of \( \sim 10 \) K using materials such as GaAs and CdTe. However, using other materials—such as GaN, ZnO and organic semiconductors—polariton condensates at higher temperatures, including room temperature, have also been realized¹³–¹⁷. This makes exciton–polaritons a fascinating topic not only from a fundamental perspective but also of potential practical interest to future quantum technological devices. As will be discussed in more detail, exciton–polariton condensates form in effectively two-dimensional structures rather than three dimensions. This allows the investigation of interesting physics peculiar to two dimensions, such as the Berezinskii–Kosterlitz–Thouless (BKT) transition, where there is an interplay of long-range order and thermally excited vortices.

In this review, we give an overview of the recent rapid progress made in the realization and understanding of exciton–polariton condensates. As there have been several reviews¹⁸–¹⁹ focusing primarily on the fundamental aspects of exciton–polaritons, here we shall emphasize some of the more recent developments relating to exciton–polariton condensates. In particular, we will discuss physical phenomena exhibited such as superfluidity, vortex formation, nontrivial phases of polariton condensates exhibiting BKT and Bardeen–Cooper–Schrieffer (BCS) physics, and the physics and applications of engineered polariton structures. One important issue which arises invariably when discussing exciton–polaritons is the role played by the open-dissipative nature of the system. From the early investigations of coherence formation in polaritons it has been a controversial issue as to whether the concept of an exciton–polariton BEC is valid at all from a thermodynamical perspective. We will discuss the known differences between a standard photon laser, a polariton laser and a polariton BEC, and discuss how the non-equilibrium character of the system changes the nature of the physical effects observed.

Exciton–polaritons: basic aspects

We first describe briefly some basic aspects of exciton–polaritons. For a more detailed exposition we refer the reader to reviews

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such as refs 18–24. We shall primarily give examples for the most widely used GaAs- and CdTe-based systems for the sake of concreteness, although other materials are conceptually similar but with different parameter values. A typical planar microcavity for exciton–polaritons consists of several quantum wells (QWs) sandwiched by two distributed Bragg reflectors (DBRs), as shown in Fig. 1a. A QW is a thin layer (typically of the order of 10 nm) of a relatively narrow bandgap material (such as GaAs or CdTe) surrounded by a wider bandgap material (doped with Al or Mg respectively). The QW exciton is the primary excitation of the system under external stimulation; for example, by illumination from a laser. An exciton is a bound electron–hole pair originating from their mutual Coulomb attraction. The excitons are strongly confined in the growth (z-) direction owing to the QW but are free to move in the x–y plane. Excitons, by virtue of being composite particles made of two fermions, obey bosonic statistics as long as their density is low enough such that they do not overlap (the typical Bohr radius of a GaAs QW exciton is ~10 nm; ref. 25).

The use of several QWs allows the total exciton density to be distributed over the QWs to avoid reaching this saturation (or Mott) density, while simultaneously enhancing the cavity coupling. Excitons have typical lifetimes of the order of 100 ps to 1 ns in GaAs and CdTe semiconductors.

Although excitons already have a relatively light effective mass by atomic standards (~0.2m_e, where m_e is the bare electron mass), by using a cavity it is possible to make them considerably lighter\(^1\). Photons are confined within the cavity in the z-direction, whereas they are free to move in the x–y plane (Fig. 1a). This gives them an effective mass typically of the order ~10\(^{-4}\)m_e. The effect of the cavity is to create strong coupling between the mobile excitons and photons, resulting in new quasiparticles which are a superposition of the two: exciton–polaritons. The resulting dispersion relations are shown in Fig. 1b. The photon and exciton dispersions anticross under strong coupling, resulting in two new dispersion relations for the lower polariton (LP, lower energy branch) and upper polariton (UP, higher energy branch). The Rabi splitting \(\Omega\) of exciton–polaritons (defined as the energy difference between the LP and UP dispersions at zero in-plane momentum \(k\)) is typically of the order of 10 meV (in multi-QW GaAs and CdTe samples), and is larger than or comparable to relevant system parameters such as the temperature, binding energy and interactions. This allows us to consider to good approximation the quasiparticles of the system to be exciton–polaritons, rather than the original excitons or photons. The exciton–polariton effective mass and lifetime are predominantly determined by their photonic component, whose effective mass is much lighter and whose lifetime is much shorter than the excitonic part. The photons have a finite lifetime due to leakage of the light through the microcavity mirrors. Samples that were used in early experiments had lifetimes typically of the order of \(\tau\sim1\) ps; however, more recently, these have been extended to \(\tau\sim10–100\) ps (refs 27,28). Whereas the polariton lifetimes are of the order of the photon, the polariton–polariton interactions are inherited from their excitonic component, arising primarily from Coulomb exchange effects of the underlying fermionic species\(^29,30\). This gives a typical mean field interaction of the order of meV, resulting in extremely light quasiparticles with short lifetimes, but simultaneously possessing a sizeable nonlinear interaction.

Condensation then occurs by the following process. First a population of polaritons must be introduced, which requires excitation from an external source, usually a laser (Fig. 1b). As the aim is to show spontaneous coherence of the polaritons, they should be introduced in a way such that the original coherence of the laser is lost. This can be done primarily by two methods: resonant pumping to exciton energy at a large angle (Fig. 1b) and non-resonant pumping at the reflection minimum of the stop band at \(k=0\) (ref. 31). In either method, a hot cloud of polaritons is produced, mostly occupying the LP dispersion. The LPs then undergo scattering from the crystal and dissipate their energy via phonon emission. This cooling happens efficiently down to momenta such that the exciton and photon energy difference is of the order of the Rabi splitting, which is where the photonic component of the polariton becomes appreciable\(^32\). At this momentum the lifetime of the polariton is greatly reduced, such that less time is available for cooling. Simultaneously the dispersion steepens owing to the Rabi splitting at this momentum, reducing the phonon density of states and making the cooling less efficient. This creates a ‘bottleneck’ in the polariton population\(^33,34\), and a second mechanism becomes responsible for further cooling of the polaritons. For a sufficiently dense population of the bottleneck polaritons, polariton–polariton scattering takes over as the dominant cooling process. For example, two polaritons within the bottleneck region can scatter, leaving one in the vicinity of \(k=0\) and another at twice the bottleneck momentum. The higher energy polariton can cool again via phonon emission, resulting in a lower overall energy of the cloud. This secondary cooling mechanism allows momentum states within this bottleneck region to build up a macroscopic population. The population buildup occurs most probably at the \(k=0\) state by virtue of its lowest energy. We note that other pumping schemes in addition to the above two methods have been used in several works, most notably the optical parametric oscillator (OPO) scheme\(^35\).

Some typical measurement data of condensation, by Kasprzak, Devaud and co-workers\(^2\), are shown in Fig. 1c. Here we see the energy–momentum dispersion relation, directly measured from the photoluminescence emerging from the microcavity. Polaritons decay by leakage through the DBR mirrors, emerging as a photon carrying the same energy and momentum as the polaritons in the QWs. The energy can be measured by spectroscopy, while the momentum can be found from the angular direction of the photon propagation. Below condensation, there is a broad distribution of exciton–polaritons in both energy and momentum of the LP dispersion. As the pumping power is increased, there is a sudden narrowing in both the energy and momentum distribution and a large population of polaritons occupies the zero-momentum mode of the system, consistent with the formation of a BEC. Figure 1d plots the typical dependence of the polariton population on the pump power. Below threshold, the population at \(k=0\) increases linearly with the pump power, but increases nonlinearly with the onset of condensation\(^36\). The population dependence returns to a linear dependence after the transition region, as now the condensate population dominates the total polariton population. The macroscopic occupancy of the ground state, the nonlinear threshold behaviour, and the narrowing of the linewidth are all properties that would be expected of BEC. However, these alone are not sufficient to conclude that BEC has occurred, and one requires careful cross checking with other expected properties. This is the topic of the next section.

Is it a laser or a BEC?

One of the most controversial issues relating to exciton–polariton BECs is whether it should be called a BEC at all. Looking again at the device structure in Fig. 1a, it consists of a cavity enclosing a QW which supports electron–hole excitations—precisely the same structure as a vertical-cavity surface emitting laser (VCSEL). In the VCSEL, the electrons and holes act as the gain medium and lasing occurs via population inversion, where many electron–hole pairs are excited. Coherent light is emitted by the usual process of stimulated emission, where the light in the cavity is amplified by recombinination of electron–hole pairs. In the procedure described in the previous section for creating the polariton condensate, we excite a large population of high-energy excitons to supply the condensate. Could not the coherent light emitted by the polariton BEC be more simply...
Exciton–polariton condensation. a) Typical device structure supporting exciton–polaritons. Excitons, consisting of a bound electron–hole pair, exist within the quantum well layers. These are sandwiched by two distributed Bragg reflectors (DBRs), made of alternating layers of semiconductors with different refractive indices. The DBRs form a cavity that strongly couples a photon and an exciton to form an exciton–polariton. Polaritons are excited by a pump laser incident from above. b) Exciton–polariton dispersion and condensation process. Strong coupling between the cavity photon and exciton dispersions split the dispersions near \( k = 0 \) to create the lower polariton (LP) and upper polariton (UP) dispersions. The pump laser initially excites high-energy excitons, which then cool via phonon emission towards the bottleneck region (black clouds). We show both the resonant pumping scheme (large blue arrow) and the non-resonant pumping scheme (large red arrow, pumped at a higher energy beyond the scale shown). Excitons in the bottleneck region then scatter into the condensate (orange cloud) via stimulated cooling. c) Figure 1 Experimental dispersion images of polariton condensate formation from ref. 5. Below the threshold for condensation the polaritons are broadly distributed in momentum and energy. At and above threshold the polaritons condense in the \( k = 0 \) ground state. d) Polariton ground state population for a polariton laser as a function of the pump power from ref. 36. The figure also shows the threshold for a standard laser achieved by a sufficiently large detuning to lose strong coupling in the same sample for comparison.

interpreted as standard lasing, where the excitons play the role of the gain medium. Another question relates to the short lifetime of the polaritons, which means that the condensate must be continually replenished to have a stable population. As the usual concept of a BEC assumes thermodynamic equilibrium, in such an explicitly non-equilibrium situation, does it make sense to even think of a BEC? Although some of these issues are the subject of ongoing debate, we describe the present state of understanding addressing these questions.

The first difference between a polariton BEC and a VCSEL is which particle species accumulates coherence (Fig. 2a). From a device perspective, one clear difference between a polariton BEC and a VCSEL microcavity structure is the presence or absence of strong coupling, respectively. In the lasing case, the gain medium (electron–hole pairs) is pumped sufficiently such that population inversion occurs. Then, via a process of stimulated emission, photons are emitted and amplified coherently such that eventually lasing occurs. Thus the species that develops coherence consists of the photons and the gain medium is not coherent. In contrast, in a polariton BEC a large population of hot uncondensed polaritons is initially excited. Assuming that the polariton lifetime is sufficiently long for thermalization with the crystal to occur, by a process of stimulated scattering into the \( k = 0 \) mode, in a similar way to Fig. 1b, a polariton condensate forms. The polaritons that are in the condensate then emit coherent light via leakage of their photonic components through the microcavity mirrors. In this case, the coherence that accumulates is in the polaritons rather than the cavity photons. Thus, even though both the laser and the polariton condensate emit coherent light, in this case there is a clear distinction determined by what particle species becomes coherent. More precisely, the process of coherence formation is different owing to the different particles involved—lasers become coherent by stimulated emission, whereas polariton condensates are coherent owing to stimulated cooling.

Although the above distinction is clear if polariton lifetimes are very long relative to the thermalization time, this is not always the case in practice, making the distinction less obvious. In principle, for particles with infinite lifetimes the photonic or excitonic fractions do not matter—we may equally consider the two extremes of the spectrum of photon BECs (ref. 12) and exciton BECs. The issue of the particle lifetime brings another aspect to the discussion, of whether thermal equilibrium has been reached. In the literature it has now become commonplace to refer to different regimes in various ways. At one end of the spectrum is the polariton BEC,
which is the ideal case discussed above, where the lifetimes are long enough such that thermalization occurs. At the other end of the spectrum, strong coupling is lost, which may arise in a variety of different ways, such as short lifetimes or additional dephasing. This is referred to as a photon laser. An intermediate regime, where strong coupling and macroscopic occupation of the polariton ground state is present, but without a thermalized population of polaritons, is referred to a polariton laser. The polariton laser was originally introduced as a novel type of laser where population inversion was not necessary to form a coherent polariton population\(^{44,45}\). The term polariton condensate is used as a broader term to encompass both the polariton BEC and polariton laser.

In the early experiments the polariton lifetimes were of the order of \(\tau \sim 1\) ps and the stimulated scattering times of a similar order or less\(^{46}\). Nevertheless, thermalization of the polaritons following a Boltzmann exponential decay of the distribution function was observed at threshold (Fig. 2b). At higher excitation powers, there is an increase in population of particles at lower energy, deviating from the exponential distribution, as would be expected of a BEC. Whereas in an ideal BEC one would expect that the population follows a Bose–Einstein distribution, for polariton condensates the fit to such a distribution is found to be generally rather poor. Apart from technical issues such as the difficulty in estimating the chemical potential and polariton temperature, the presence of uncondensed excitons, density and non-equilibrium effects are all contributing factors that significantly alter the distribution. In this respect it may not be surprising that the Bose–Einstein distribution is not followed precisely. Experiments showing condensation are performed at detunings such that the excitation fraction is larger than the photon fraction, which reduces the cooling time and extends the lifetime, favouring condensation. Increasing the photonic component enables one to shift the condensate away into a more distinctly non-equilibrium regime\(^{45}\). It has been argued that the fact that polariton condensates can form without thermal equilibrium suggests it should be more appropriately called a polariton laser\(^{19}\). These results suggest that experiments at present have just entered the regime where equilibrium can be discussed, but is not very far across the boundary. As the quality of the samples improves and polariton lifetimes are extended, it is reasonable to assume that polariton condensates that are increasingly closer to thermal equilibrium could be achieved.

Thermalization is just one of many characteristics that is expected in a BEC. Table 1 gives a checklist of characteristics for a polariton BEC, a polariton laser and a photon laser. For a BEC off-diagonal long-range order is a central concept, where there is the appearance of a macroscopic wavefunction \(\psi(r)\) forming an order parameter\(^2\). Experimentally this corresponds to extended spatial coherence \(g^{(1)}(r)\) across the condensate above threshold, but short-range correlations below threshold—a fact confirmed in experiments such as refs 5,47. In addition to the spatial coherence, the polaritons should occupy one of the spin degrees of freedom macroscopically (optically active polaritons have spin \(\pm 1\)), which is observed as a linear polarization of the emitted light above the condensation threshold\(^2,16\). For cases where there is no bias in any spin direction, the polarization emerges stochastically, and the spin symmetry is spontaneously broken\(^{13}\).

For a macroscopic wavefunction, the position and momentum uncertainty product is of the order of the Heisenberg limit, as confirmed in ref. 47. Other effects that are expected are an increase in temporal coherence, originating from the spectral narrowing of the condensate, and confirmation that condensed particles are polaritons as opposed to cavity photons. As discussed above, the detection of whether the coherent particle species is a polariton or a photon is at the heart of distinguishing between various types of coherence. However, spectroscopic experimental data do not always provide unambiguous evidence, as several contributing factors may affect the energy of the polariton. In such cases the measurement of a Zeeman shift by an externally applied magnetic field has been used to identify the existence of an excitonic component to the polaritons, distinguishing it from a photon laser\(^{48}\).

Despite progress in the realization and understanding of a polariton BEC, questions remain. The buildup of spontaneous coherence between the two extremes of the polariton BEC and photon laser\(^{49}\) suggests that it may be more appropriate to think in a unified way of the continuum between a laser, a distinctly non-equilibrium phenomenon, and an equilibrium BEC. To investigate this aspect there has been increased activity in the identification of a second threshold above the polariton BEC transition. At pumping powers considerably above the condensation threshold (typically about 10–100 times), observations that seem to be photon lasing have been known since early investigations of polariton coherence\(^{44,50}\). Theoretically the mechanism of this is now only beginning to be understood\(^{51}\). Another question relates to the steady leakage and replenishment of the polaritons to maintain a constant condensate population. This potentially changes the properties of the BEC, such as superfluidity and its various quantum phases, in a fundamental way. How do we understand the physics of BECs in this new non-equilibrium setting? Some of these aspects will be discussed in the following sections, but are an ongoing topic of investigation.

### Superfluidity

Superfluidity is the phenomenon of flow without friction, and is often observed in systems that undergo spontaneous coherence, such as BECs (ref. 2), liquid \(^4\)He (ref. 3) and superconductors\(^4\). It allows the existence of spectacular effects, such as persistent

**Table 1** Differences between an exciton–polariton BEC, exciton–polariton laser, and a vertical cavity surface emitting laser (VCSEL).

| Property                        | Exciton–polariton BEC | Exciton–polariton laser | VCSEL |
|---------------------------------|-----------------------|-------------------------|--------|
| Thermal equilibrium below threshold\(^{3,6}\) | ✓                     | x                       | x      |
| Bose distribution above threshold | ✓                     | x                       | x      |
| Threshold corresponds to onset of degeneracy\(^{5,7}\) | ✓                     | ✓                       | x      |
| Linewidth narrowing\(^{3,6}\) | ✓                     | ✓                       | x      |
| Increase of temporal coherence \(g^{(1)}(r)\)^{5,113} | ✓                     | ?                       | ✓      |
| Spontaneous polarization\(^4\) | ✓                     | ✓                       | x      |
| Long-range spatial coherence \(g^{(1)}(r)\)^{5,6,47} | ✓                     | ✓                       | x      |
| Polaritons are the particles that accumulate coherence (strong coupling)\(^{48}\) | ✓                     | ✓                       | x      |
| Heisenberg–limited position and momentum uncertainty product\(^{47}\) | ✓                     | ✓                       | x      |

The properties that have been experimentally demonstrated for polariton condensates are shown as the references.
currents that flow indefinitely, the impossibility of rotating the superfluid, and flow through capillaries without viscosity. Central to the concept of superfluidity is the Landau criterion, which gives the maximum velocity \( v_c = \min_k \epsilon_k/k \) at which the fluid can flow while still maintaining its superfluid properties, where \( \epsilon_k \) is the energy-momentum dispersion. Although the Landau criterion is typically derived from general principles of Galilean transformations, it is also possible to understand its origins directly from a simple scattering argument. For a moving condensate with a parabolic dispersion, particles within the condensate always have other states with the same energy to scatter into. The scattering eventually destroys the single-momentum state occupation that the condensate was originally in, and eventually acts to slow the average motion. However, if the moving condensate has a linear dispersion arising from Bogoliubov interactions, and if the velocity is less than the Landau velocity, then there are no resonant states to scatter into. This suppression of scattering then acts to maintain the condensate velocity, even in the presence of scattering defects, resulting in superfluidity.

How well does this argument transfer to polariton condensates? Again, the non-equilibrium nature makes the straightforward application of this argument problematic. Experimentally, evidence exists showing polariton condensates possess a linear Bogoliubov dispersion. However, in the more general case it is widely believed that the excitation spectrum is modified owing to the open-dissipative nature of the condensate. The model, introduced by Wouters and Carusotto, which is often used to describe the system is a dissipative Gross–Pitaevskii (GP) equation with gain and loss terms, coupled to an incoherent reservoir. The reservoir is a dynamical variable, which can modify the dispersion of the condensate rather strongly. The energies of the dissipative GP eigenstates also become complex in general, reflecting the decaying nature of the excitations. A typical excitation spectrum for a homogeneous condensate potential is shown in Fig. 3a. Instead of the lowest-lying excitations starting linearly around \( k = 0 \), they instead begin only at a non-zero value. Thus the main characteristic of the Bogoliubov dispersion, the linearity around zero momentum, is not necessarily present in polariton BECs (although for particular parameters it can exist). The problem is that when the Landau criterion is applied to such a dispersion, it always gives \( v_c = 0 \), implying that superfluidity never occurs. However, this naive application of the Landau criterion contradicts numerical evidence showing suppressed Rayleigh scattering, persistent current flow and suppressed drag. Figure 3b shows a numerical calculation of condensate flow at various velocities in the presence of a defect. We see that, despite the dissipative nature of the GP equation, there is a characteristic velocity at which the fringes due to the defect disappear, equal to the sound velocity (the gradient of the Bogoliubov dispersion at \( k = 0 \)).

The reservoir \( g |\psi|^2/m \), where \( g \) is the interaction energy, \( |\psi| \) is the density and \( m \) is the polariton mass. This occurs even despite the lack of any definable Landau velocity in the dispersion. Whereas the dispersion gives a negative prediction relating to superfluidity in dissipative systems, an analysis based on Green's functions gives a different conclusion, with the presence of a normal and superfluid component. On the other hand, calculations based on the superfluid stiffness show that superfluidity does not survive in the thermodynamic limit, although for finite systems superfluid behaviour can be present.

From an experimental point of view, several works have shown evidence consistent with superfluidity in exciton–polariton condensates. The most compelling evidence of superfluidity so far is the measurement of Rayleigh scattering of a polariton condensate in the presence of an impurity. In the experiments performed by Amo and co-workers, a polariton condensate is excited by a continuous wave laser, and centred around natural defects that exist in the sample. As shown in Fig. 3c, as the density is increased the amount of Rayleigh scattering due to the condensate flow diminishes. This is attributed to an increase in the critical velocity according to the Landau criterion, as the sound velocity \( c_s \) increases with density. At low density, the distribution of the polaritons in momentum space has strong scattering to momenta of equal magnitude, a clear signal of Rayleigh scattering, whereas at higher densities the scattering is suppressed, as would be expected in a superfluid. In the experiments of Sanvitto and co-workers, first a polariton condensate is prepared, then a 2 ps pulsed laser in a Laguerre–Gauss mode carrying angular momentum is imprinted on the condensate to induce a vortex state. The circular flow induced by the vortex was seen to survive for long times, limited only by the uncontrollable random walk within the condensate which the vortex core undergoes. Although these experimental results have been interpreted in terms of superfluidity, it seems to have been pointed out that this may not be the only way to understand the results. Butov and Kovkin have led the opposing view that the suppression of scattering could originate from other mechanisms, such as screening of the disorder potential and large propagation distances due to the fast polariton velocities within the experimental lifetime.
Figure 3 | Superfluidity in exciton–polariton condensates. a, Excitation spectrum of the dissipative Gross–Pitaevskii (GP) equation. The bare lower polariton dispersion (no interaction or dissipation) and the Boguliubov dispersion are shown with the dashed lines for comparison. Momentum is normalized to the decay momentum $k_0 = \sqrt{m/\hbar}$ and energy $E_0 = \hbar^2 k_0^2 / 2m$. Typical numbers are, for example, $k_0 = 1 \mu m^{-1}$ and $E_0 = 0.68$ meV. The parameters used are the same as in Fig. 1a of ref. 56. b, Numerical simulation of the density of a moving condensate in real space with a single defect using the dissipative GP equation in ref. 58. The velocity of the condensate is (i) $v/c_\text{y} = 1.5$, (ii) $v/c_\text{y} = 1$, (iii) $v/c_\text{y} = 0.4$. c, Experimental real (top row) and momentum (bottom row) space images of a polariton condensate impacting on a defect in ref. 61. As the velocity of the condensate is reduced (left to right), in real space the fringes disappear, while in momentum space Rayleigh scattering becomes suppressed.

The mixed results in theory and experiment suggest that further work is required to completely understand the notion of superfluidity in this non-equilibrium setting. Another question regarding the excitation spectrum is the observation of the `ghost' branch of the Bogoliubov spectrum, which has been predicted to exist theoretically\textsuperscript{58}. In addition to the standard positive Bogoliubov spectrum, a negative branch should be visible in the photoluminescence. This was observed in the OPO regime\textsuperscript{59}, but so far there has been no direct photoluminescence measurement.

Phases of polariton condensates

In an infinite two-dimensional (2D) system, off-diagonal long-range order associated with BEC in a non-interacting quantum degenerate Bose gas breaks down because it is vulnerable to thermal fluctuations at non-zero temperatures\textsuperscript{60,61}. However, a 2D interacting Bose gas in an infinite system is predicted to exhibit quasi-long-range order, preserving superfluidity by spontaneously forming vortex pairs via the BKT transition\textsuperscript{62,63}. Above the BKT critical temperature, the quantum gas excites free single vortices induced by phase fluctuations. However, when the system cools down, the vortices pair up and phase fluctuations in space cancel out, reducing the free energy, and recovering quasi-long-range coherence and superfluidity. BKT physics has been one of the central themes in the investigation of low-dimensional quantum fluids in recent years\textsuperscript{64}. In particular, there is still an incomplete understanding of the quantum phase of an interacting Bose gas in a finite-sized 2D system. Exciton–polariton superfluids are a good testing ground to examine this unresolved problem, and active theoretical and experimental efforts have examined the gain–loss mechanism and the dynamics of vortices via time-resolved and phase-resolved imaging techniques.

A quantized vortex is a topological defect with zero density at its core and a multiple of $2\pi$ phase rotation around it. Single vortices and vortex pairs in exciton–polariton condensates excited by non-resonant pumping have been observed either pinned at defects or imprinted from the phase fluctuation of lasers. Vortices are detected via interferometry, where the spatial phase and intensity distributions are reconstructed. The first observation of vortices in exciton–polaritons was reported for CdTe systems\textsuperscript{65}. In this experiment, Lagoudakis and co-workers identified a fork-like interference pattern, which is the signature of a phase dislocation at an accidental local potential minimum (Fig. 4a). The same team also observed pinned half-quantum vortices using polarization-resolved interferograms, which show a $\pi$-rotation in phase and a $\pi$-rotation in polarization\textsuperscript{71}. Figure 4b shows the image of a single vortex–antivortex pair observed in a GaAs system, induced by inhomogeneities in the spot profile of the pump laser\textsuperscript{72}. The disassociation of a single vortex in a spinor polariton condensate into half-vortices has also been observed\textsuperscript{73}. Whereas all the above works are under an incoherent pumping scheme, vortex–antivortex (V–AV) pairs have also been observed in the OPO regime\textsuperscript{74}. Experimental observations have been consistent with numerical calculations using a dissipative GP equation, although it should be emphasized that the existence of vortices is not evidence for a BEC—vortices can, for example, exist in optical and charge-density-wave systems.

Although the observation of vortices and V–AV pairs may suggest a connection to a BKT phase, in fact experiments at present are not yet conclusive on this. In a true BKT phase, vortices and V–AV pairs associated with phase fluctuations are thermally activated. One way to identify the BKT phase would be via a single-shot measurement, but the low signal-to-noise ratio...
of short-lived polaritons is at present a limitation. Another way to explore the BKT phase is to characterize the spatial correlation function $g^{(1)}(r)$, which quantifies the off-diagonal long-range order of the system. The BKT phase is predicted to show a power-law decay of $g^{(1)}(r)$, exhibiting quasi-long-range spatial coherence. This is possible if bound V–AV pairs are created where the phase disturbance of a vortex is exactly cancelled out with that of an antivortex. Thermodynamically, V–AV pairs are favourable for decreasing the free energy by increasing the entropy. Although quantitative measurements of $g^{(1)}(r)$ in quantum fluids are lacking, exciton–polariton systems are promising to establish this directly by interferometric measurements. Several reports of $g^{(1)}(r)$ in one-dimensional exciton-polaritons\(^{75,76}\) indicate that long-range order exists above threshold. Although a direct prediction of $g^{(1)}(r)$ for the non-equilibrium polariton system is lacking, it is known from calculations based on lattice models at equilibrium that the long-range power-law decay should have an exponent of 0.25. Experimental results of exciton–polariton condensates in 2D have been reported as observing the power-law decay behaviour\(^{77,78}\), which may be indirectly attributed to the formation of bound V–AV pairs. However, smoking-gun experiments to elucidate the BKT–BEC crossover would be the single-shot direct observation of free vortices above the BKT temperature, bound V–AV pairs in the BKT phase, and a vortex-free regime at lower temperatures in the BEC phase. Whereas short-lived polaritons are unlikely to show this evidence conclusively, the recent advent of long-lived polaritons ($\tau \sim 10–100$ ps; refs 27,28) provides good candidates for showing this crossover and superfluidity.

Solitons are another type of topological excitation present in polariton BECs. Repulsive polariton–polariton interactions are predicted to support dark solitons\(^9\), which are characterized by a phase slip in the propagation of exciton–polariton fluids. Such solitary waves may be produced by disturbing the condensate with a localized obstacle. By means of an interferogram, the phase along the soliton trajectory was mapped as shown in Fig. 4c\(^{80}\). Since the first report of the observation of exciton–polariton solitons, the temporal dynamics of dark solitons\(^41\) and bright solitons at the lower-polariton inflection with negative mass\(^2\) have been observed. More recently, it was shown that polaritons in the linear regime (that is, with no polariton–polariton interactions) could also reproduce the intensity and phase patterns that are expected of solitons purely from interference effects\(^81\). The question as to what is necessary for the unambiguous claim of observing a soliton is a topic of current debate.

Another type of crossover that has been considered for polariton condensates is the BEC–BCS crossover. For exciton BECs, it has long been theoretically predicted that such a crossover should exist as the density is varied\(^{34,85}\). The physical picture of this crossover is that at low densities the electrons and holes within the exciton are relatively strongly bound by their mutual Coulomb attraction, and condense at sufficiently low temperatures owing to their bosonic nature. At higher densities, their mutual attraction becomes screened by the large population of electrons and holes, forming loosely correlated Cooper pairs, described by a BCS wavefunction. Although this picture is well established for exciton BECs, does the same hold for polariton BECs? This question was first examined by Keeling, Littlewood and co-workers\(^{86,87}\) and followed up by several others using a BCS wavefunction approach\(^{36,89}\). The consistent picture which emerges from these studies—all of which assume thermal equilibrium—is that at high densities the photons completely dominate the dynamics, in what can be described as a photon BEC state\(^{82}\). The reason for this is simple: the electrons and holes have a maximum allowable population due to the Pauli exclusion principle, as they are fermions, whereas photons do not. Therefore, at some
point the number of photons overwhelms the excitons such that the photons dominate the properties of the system. The photons have the effect of producing an effective attractive interaction induced by the Rabi coupling between the electrons and the holes. Therefore, increasing the density in polariton condensates acts generally in the opposite direction to the exciton BEC–BCS crossover: the attraction between the electrons and the holes in the excitons is reinforced by the Rabi coupling. These studies show that the presence of the photons considerably alters the quantum state of the system at sufficiently high densities, even without taking into account non-equilibrium effects. Taking the finite lifetime into account, it is expected that with a sufficient short photon lifetime all these regimes are more suitably thought of as a laser, giving a crossover between photon BEC, polariton BEC, electron–hole BCS (Cooper pairs of electrons and holes) and photon lasing regimes. The full understanding of this phase diagram is at present an active topic of theoretical and experimental research\(^{51,89,96}\).

**Engineered polariton structures**

The realization of the optical lattice in cold atomic gases has allowed an unprecedented ability to manipulate BECs, with seminal experiments such as the superfluid–Mott insulator transition\(^{81}\) kicking off the field of quantum simulations\(^{82}\). Likewise, the manipulation of the quantum state of the exciton–polariton condensate is a important task that is necessary both for investigating fundamental physics such as the BKT transition and realizing future polaritonic quantum devices. Figure 5a shows a zoo of engineering methods to create static and dynamical in-plane potentials by modifying either the photonic or excitonic modes, which consequently affects the polariton states. The techniques include chemical etching for forming pillars, strips and 2D lattices\(^{26,75,93,114}\), laser pump spot formation\(^{94}\), piezoelectric acoustic lattices\(^{95,115}\) and thin metal film deposition\(^{96,97}\). Single-trap configurations have realized energy separation of the s-orbital ground state from higher-orbital states\(^{89,99}\). Other geometries that have been realized include coupled molecules forming bonding and antibonding states\(^{100}\), and one-\(^{28,98}\) and two-dimensional lattices\(^{77,101–103}\) exhibiting band structures. For one-dimensional lattices, condensation in the excited p-orbital band has been realized\(^{96}\), a similar effect to that known in weak links of superfluid \(^{3}He\) forming metastable p-states\(^{89}\). Owing to the negative effective mass and the repulsive interparticle interaction at the Brillouin zone edge, a bright soliton was observed inside the gap, as shown in Fig. 5b\(^{28}\). The excited state condensation in lattices results from the interplay of gain-relaxation dynamics and the finite lifetime of the polaritons, making it more favourable to condense in these states. The same physics applies to 2D lattices, where the metastable d\(_{xy}\)-orbital condensation has been observed in a square lattice\(^{97}\). Other geometries, such as the honeycomb lattice\(^{103}\), have been realized, where V–AV lattices have been observed, as well as a linear dispersion due to Dirac points in triangular lattices\(^{102}\). Figure 5c shows the Brillouin zones of a honeycomb lattice potential above the polariton condensation threshold. Laser pump spot engineering can control the condensate wavefunction to produce 2D lattices, where a stable V–AV honeycomb lattice has been observed (Fig. 5d)\(^{94}\).

The creation of periodic potentials suggests applications to quantum simulation, where complex quantum many-body systems are realized which are beyond the reach of computer simulations\(^{82}\). Another possibility in this direction is to realize exotic Hamiltonians for the creation of metamaterials that have no natural realization. For instance, an interesting open question is to understand the system order when coherent p- and d-states of exciton–polaritons are hybridized. Furthermore, by incorporating spin dynamics controllable by light polarization, exciton–polaritons have the potential to explore magnetic order, which could provide insights.

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**Figure 5** | Methods of creating trapping potentials for exciton–polariton condensates. **a**, Static and dynamical potential engineering methods: etching (first and second from left, taken from ref. 114 and ref. 99, respectively), surface acoustic waves (third; ref. 115), strain (fourth; ref. 6) and the metal-film technique (rightmost). SAW, surface acoustic wave; IDT, interdigitated transducer. **b**, Experimentally observed one-dimensional band structure from a wire configuration in ref. 28. **c**, First three Brillouin zones measured for two-dimensional (2D) honeycomb lattices using the metal film technique. The scale bar indicates the size of the first Brillouin zone. **d**, 2D vortex-antivortex lattice formation using laser spot engineering from ref. 94. Location of the vortices (red crosses) and antivortices (yellow crosses) are marked.
into the behaviour of fermions in strongly correlated systems. Other types of applications include polaritronic circuits (the polariton analogue of atomtronic circuits)\(^{105-109}\) and novel light sources\(^{41,110}\). The recent achievement of electrical pumping, replacing the laser pumping methods described here, gives the potential for ultralow

threshold lasers\(^{41,48,111}\).

**Outlook**

We have seen that exciton–polariton condensates exhibit a rich variety of phenomena, possessing characteristics in common with atomic BECs, but in a previously unexplored non-equilibrium regime. Even the most fundamental question as to whether it is a BEC or a laser forces us to reanalyse what we mean exactly when we make these distinctions. As we suggested at the beginning of this review, the phenomenon of spontaneous coherence seems to be a rather robust concept, stretching between the superficially distinct quantum states of superconductors, lasers and BECs. Perhaps what exciton–polariton condensates teach us is that a generalization is required to unify these concepts together, rather than their categorization into separate regimes. What is fascinating is that, even without complete equilibration, polariton condensates exhibit many of the phenomena that are associated with BECs and lasers. We have seen that the cavity photon loss and the quasi-bosonic nature of the exciton modify familiar physical effects such as superconductivity, vortex formation, and BKT and BCS phases, giving the opportunity to re-examine these from a new perspective\(^{12}\).

As discussed throughout this review there are many open problems that are still ongoing topics of investigation. We have seen that the mechanism of non-equilibrium superfluidity, two-threshold crossover to photon lasing, evidence of BKT and BCS physics, and signatures of solitons are some of the growing list of questions being further investigated at present. Perhaps one of the most fascinating prospects this system possesses is the ability to engineer the microcavity configuration by nanofabrication. This suggests applications in quantum simulation, where quantum lattice models can be investigated in an engineerable system. Many of the fabrication methods allow, in principle, an arbitrary device geometry, not limited to periodic or simple trap configurations, and technological improvements in the quality of the samples will undoubtedly continue to occur. This control in device structure combined with room-temperature operation makes polariton condensates attractive for future quantum technological applications. Although at present it is still unclear what the major application of polariton condensates will be, the fundamental questions that they open will undoubtedly give us a better understanding of the phenomenon of spontaneous coherence, and the remarkable physics which comes with it.

Received 27 January 2014; accepted 25 September 2014; published online 31 October 2014; corrected online 13 November 2014

**References**

1. Sargent, M., Scully, M. O. & Lamb, W. E. *Laser Physics* (Addison-Wesley, 1978).
2. Pitaevskii, L. & Stringari, S. *Bose–Einstein Condensation* (Oxford Science Publications, 2003).
3. Tilley, D. B. & Tilley, J. *Superfluidity and Superconductivity* (IOP Publishing, 1990).
4. Leggett, A. *Quantum Liquids: Bose Condensation and Cooper Pairing in Condensed Matter Systems* (Oxford Univ. Press, 2006).
5. Kasprzak, J. et al. *Bose–Einstein condensation of exciton polaritons*. *Nature* 443, 409–414 (2006).
6. Balili, R., Hartwell, V., Snoke, D., Pfeiffer, L. & West, K. *Bose–Einstein condensation of microcavity polaritons in a trap*. *Science* 316, 1007–1010 (2007).
7. Deng, H., Weihs, G., Santori, C., Bloch, J. & Yamamoto, Y. *Condensation of semiconductor microcavity exciton polaritons*. *Science* 298, 199–202 (2002).
8. Anderson, M. H., Enscher, J. R., Matthews, M. R., Wieman, C. E. & Cornell, E. A. *Observation of Bose–Einstein condensation in a dilute atomic vapor*. *Science* 269, 198–201 (1995).
9. Davis, K. et al. *Bose–Einstein condensation in a gas of sodium atoms*. *Phys. Rev. Lett.* 75, 3969–3973 (1995).
10. Nikuni, T., Oshikawa, M., Oosawa, A. & Tanaka, H. *Bose–Einstein condensation of dilute magnons in TlCuCl*. *Phys. Rev. Lett.* 84, 5868–5871 (2000).
11. Demokritov, S. O. et al. *Bose–Einstein condensation of quasi-equilibrium magnons at room temperature under pumping*. *Nature* 443, 430–433 (2006).
12. Klaers, J., Schmitt, F., Weisinger, F. & Weitz, M. *Bose–Einstein condensation of photons in an optical microcavity*. *Nature* 468, 545–548 (2010).
13. Christopoulos, S. et al. *Room-temperature polariton lasing in semiconductor microcavities*. *Phys. Rev. Lett.* 98, 126405 (2007).
14. Baumberg, I. J. et al. *Spontaneous polarization buildup in a room-temperature polariton laser*. *Phys. Rev. Lett.* 101, 136408 (2008).
15. Kéné-Cohen, S. & Forrest, S. R. *Room-temperature polariton lasing in an organic single-crystal microcavity*. *Nature Photon.* 4, 371–375 (2010).
16. Guillet, T. et al. *Polariton lasing in a hybrid bulk ZnO microcavity*. *Appl. Phys. Lett.* 99, 161104 (2011).
17. Plumhof, J. D., Stöferle, T., Mai, L., Scherf, U. & Mahrt, R. F. *Room-temperature Bose–Einstein condensation of cavity exciton–polaritons in a polymer*. *Nature Mater.* 13, 247–252 (2014).
18. Deng, H., Haug, H. & Yamamoto, Y. *Exciton–polariton Bose–Einstein condensation*. *Rev. Mod. Phys.* 82, 1489–1537 (2010).
19. Kavokin, A. *Exciton–polaritons in microcavities*. Recent discoveries and perspectives. *Phys. Status Solidi B* 247, 1898–1906 (2010).
20. Richard, M. et al. *Exciton–polariton Bose–Einstein condensation: Advances and issues*. *Int. J. Nanotech.* 7, 688–683 (2010).
21. Smoke, D. & Littlewood, P. *Polariton condensates*. *Phys. Today* 63, 42–47 (2010).
22. Keeling, I. & Berloff, N. G. *Exciton–polariton condensation*. *Contemp. Phys.* 52, 131–151 (2011).
23. Timofeev, V. & Sanvitto, D. (eds) *Exciton Polaritons in Microcavities* Vol. 172 (Springer, 2012).
24. Carusotto, I. & Ciuti, C. *Quantum fluids of light*. *Rev. Mod. Phys.* 85, 299–366 (2013).
25. Yamamoto, Y. & Imamoglu, A. *Mesoscopic Quantum Optics* (John Wiley and Sons, 1999).
26. Weisbuch, C., Nishioka, M., Ishikawa, A. & Arakawa, Y. *Observation of the coupled exciton–photon mode splitting in a semiconductor quantum microcavity*. *Phys. Rev. Lett.* 69, 3314–3317 (1992).
27. Nelsen, B. et al. *Dissipationsless flow and sharp threshold of a polariton condensate with long lifetime*. *Phys. Rev. X* 3, 041015 (2013).
28. Tanese, D. et al. *Polariton condensation in solitonic gap states in a one-dimensional periodic potential*. *Nature Commun.* 4, 1749 (2013).
29. Schmitt-Rink, S., Chana, P., Knight, D. & A. *Theory of transient excitonic optical nonlinearities in semiconductor quantum-well structures*. *Phys. Rev. B* 32, 6601–6609 (1985).
30. Ciuti, C., Savona, V., Piemarocchi, C., Quattropani, A. & Schwendimann, P. *Role of the exchange of carriers in elastic exciton–exciton scattering in quantum wells*. *Phys. Rev. B* 58, 7926–7933 (1998).
31. Del Valle, E. et al. *Dynamics of the formation and decay of coherence in a polariton condensate*. *Phys. Rev. Lett.* 103, 096404 (2009).
32. Tassone, F. & Yamamoto, Y. *Exciton–exciton scattering dynamics in a semiconductor microcavity and stimulated scattering into polaritons*. *Phys. Rev. B* 59, 10830–10842 (1999).
33. Pau, S., Björk, G., Jacobson, J., Cao, H. & Yamamoto, Y. * Stimulated emission of a microcavity dressed exciton and suppression of phonon scattering*. *Phys. Rev. B* 51, 7090–7100 (1995).
34. Tassone, F., Piemarocchi, C., Savona, V., Quattropani, A. & Schwendimann, P. *Bottleneck effects in the relaxation and photoluminescence of microcavity polaritons*. *Phys. Rev. B* 56, 7554–7563 (1997).
35. Spano, R. et al. *Coherence properties of exciton polariton OPO condensates in one and two dimensions*. *New J. Phys.* 14, 075018 (2012).
36. Deng, H., Weihs, G., Snoke, D., Bloch, J. & Yamamoto, Y. *Polariton lasing vs. photon lasing in a semiconductor microcavity*. *Proc. Natl Acad. Sci. USA* 100, 15318–15323 (2003).
37. Kira, M. et al. *Quantum theory of nonlinear semiconductor microcavity luminescence explaining "Boser" experiments*. *Phys. Rev. Lett.* 79, 5170–5173 (1997).
38. Butov, L. V. *A polariton laser*. *Nature* 447, 540–541 (2007).
39. Butov, L. V. & Kavokin, A. V. *The behaviour of exciton–polaritons*. *Nature Photon.* 6, 2 (2012).
40. Devaeu, P. & Proulx, R. B. *The behaviour of exciton–polaritons*. *Nature Photon.* 6, 205 (2012).
105. Deveaud-Plédran, B. Polaritronics in view. Nature 453, 297–298 (2008).
106. Liew, T. C. H., Kavokin, A. & Shelykh, I. A. Optical circuits based on polariton neurons in semiconductor microcavities. Phys. Rev. Lett. 101, 016402 (2008).
107. Amo, A. et al. Exciton–polariton spin switches. Nature Photon. 4, 361–366 (2010).
108. Nguyen, H. S. et al. Realization of a double-barrier resonant tunneling diode for cavity polaritons. Phys. Rev. Lett. 110, 236601 (2013).
109. Ballarini, D. et al. All-optical polariton transistor. Nature Commun. 4, 1778 (2013).
110. Byrnes, T., Yamamoto, Y. & van Loock, P. Unconditional generation of bright coherent non-Gaussian light from exciton–polariton condensates. Phys. Rev. B 87, 201301(R) (2013).
111. Bhattacharya, P., Xiao, B., Das, A., Bhownick, S. & Heo, J. Solid state electrically injected exciton–polariton laser. Phys. Rev. Lett. 110, 206403 (2013).
112. Snoke, D. A feature rather than a bug. Nature Phys. 4, 673 (2008).
113. Love, A. P. D. et al. Intrinsic decoherence mechanisms in the microcavity polariton condensate. Phys. Rev. Lett. 101, 067404 (2008).
114. Bajoni, D. et al. Polariton laser using single micropillar GaAs–GaAlAs semiconductor cavities. Phys. Rev. Lett. 100, 047401 (2008).
115. De Lima, M. M. Jr et al. Phonon-induced polariton superlattices. Phys. Rev. Lett. 97, 045501 (2006).

Acknowledgements
We thank B. Deveaud-Plédran for providing valuable comments on the manuscript. This work is supported by the FIRST program through JSPS, the Okawa Foundation, the Transdisciplinary Research Integration Center, and DARPA QuEST program through Navy/SPAWAR Grant N66001-09-1-2024, the Inamori Foundation, NTT Basic Laboratories and JSPS Kakenhi Grant Number 26790061.

Author contributions
T.B. and N.Y.K. wrote the manuscript. Y.Y. oversaw the work.

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Competing financial interests
The authors declare no competing financial interests.
In the version of this Review Article originally published, the sources of two images in Fig. 5a were incorrect. The first and second images from the left in Fig. 5a were taken from ref. 114 and ref. 99, respectively. This error has now been corrected in the online versions of the Review Article.