The new era of polariton condensates

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The new era of POLARITON CONDENSATES

David W. Snoke and Jonathan Keeling

Quasiparticles of light and matter may be our best hope for harnessing the strange effects of quantum condensation and superfluidity in everyday applications.

Imagine, if you will, a collection of many photons. Now imagine that they have mass, repulsive interactions, and number conservation. The photons will act like a gas of interacting bosonic atoms, and if cooled below a critical temperature, they will undergo a well-known phase transition: Bose–Einstein condensation. You will have a "superfluid of light."[1]

Now imagine that you can choose the photons' mass. Then, because the critical temperature for Bose–Einstein condensation depends on particle mass and density, you can create the condensed state even at room temperature.

That is not an idle dream. Condensed-matter physicists have a long history of inventing novel quasiparticles, such as massless electrons, particles with fractional charge, and particles with spins detached from their charges. Two decades ago researchers began to engineer hybrid particles of light and matter, called polaritons,[2,3] that could be used to realize the Bose–Einstein condensates of light described above. Today, the study of polariton condensates has come of age: Researchers have advanced beyond merely demonstrating that they exist to demonstrating ways to harness them in optical devices, including low-threshold lasers, superfluid photonic circuits, and all-optical transistors. Considerable work—both fundamental and applied—remains to turn those ideas into practical technologies. But the dream isn't as distant as it once seemed.

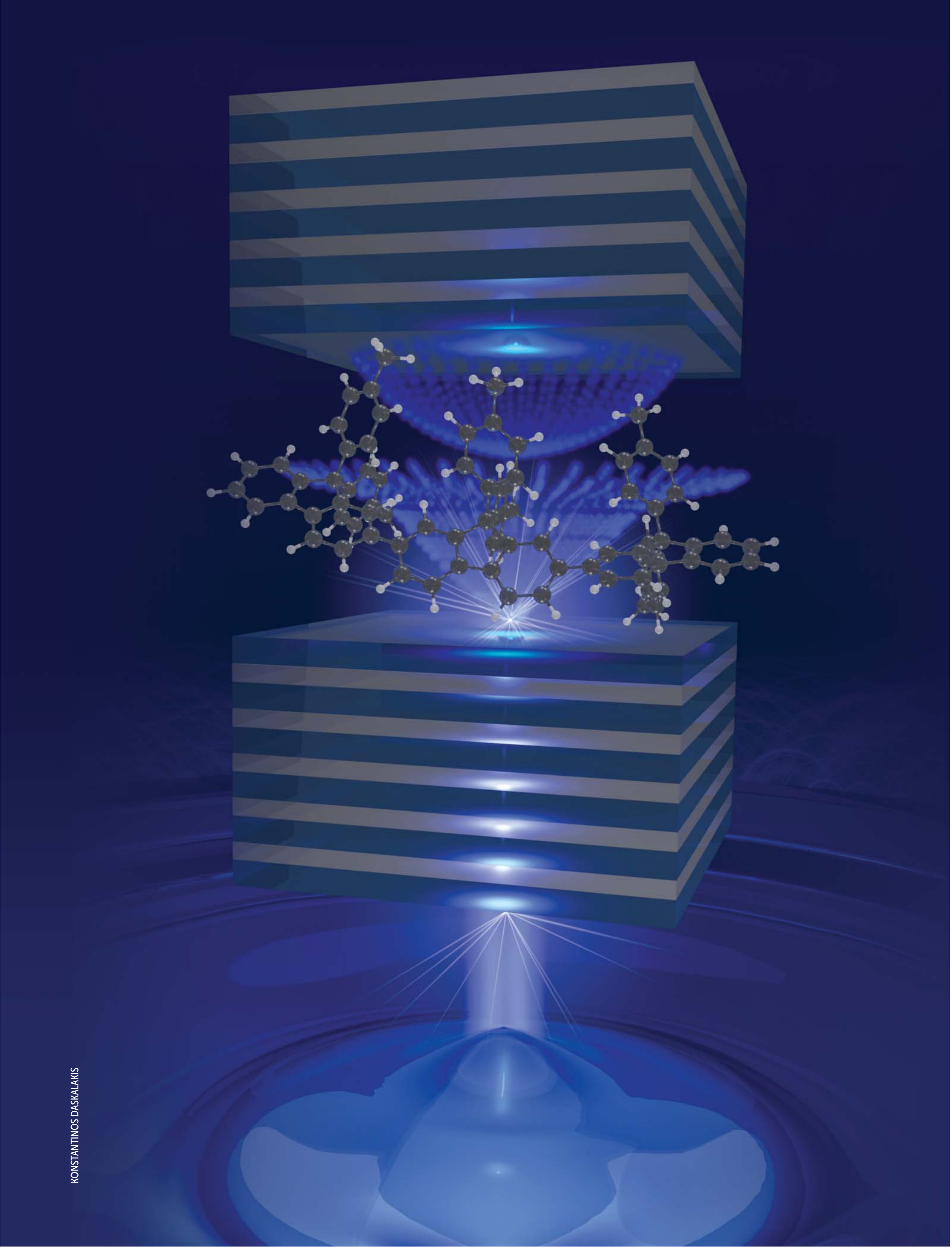
The recipe

How does one engineer a Bose–Einstein condensate from light? The first step—giving photons mass—is straightforward. In free space, photons are massless. But trapped in a cavity between two planar mirrors, as illustrated in figure 1, they acquire an in-plane dispersion relation indicative of an effective mass. That relation is given by

$$\hbar\omega = \hbar(c/n)\sqrt{(N\pi/L)^2 + k_\parallel^2},$$

where c is the speed of light, n is the refractive index inside the cavity, N is an integer corresponding to the cavity mode, L is the distance between the mirrors, and $k_\parallel$ is the in-plane wavevector. For small $k_\parallel$, the expression has a quadratic dependence on $k_\parallel$ and can be fitted with a parabola corresponding to an effective mass m equal to $\pi\hbar Nn/Lc$.

The optical cavity also addresses the issue of number conservation: In a high-quality cavity, photons can reflect off the mirrors many thousands of times before escaping. If during that time the photons equilibrate to a well-defined temperature, one can consider them, for all practical purposes, to be conserved. Because even the best cavities leak, however, the cavity



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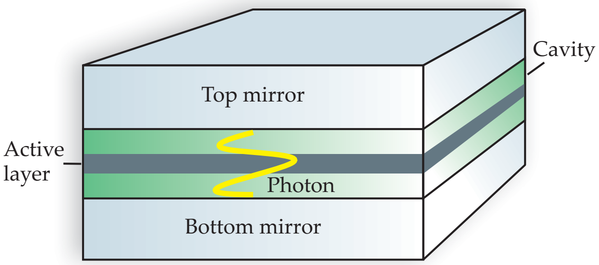

FIGURE 1. A PHOTON TRAPPED BETWEEN TWO MIRRORS acquires an effective mass, approximate number conservation, and—if tuned to an electronic excitation in an active layer sandwiched between the mirrors—the ability to interact with other photons. In other words, it acquires the properties necessary to produce a Bose–Einstein condensate.

must be continually replenished with photons from outside to maintain a constant population. Number conservation isn't strictly a prerequisite for condensation: Coherence, macroscopic occupation of single-particle modes, and other features associated with Bose–Einstein condensation can be observed in partially equilibrated systems. However, full thermal equilibration is necessary for true superfluidity.

Perhaps the trickiest step in creating our Bose–Einstein condensate of light is making the photons equilibrate. One approach is to make the photons interact, so they can scatter off each other. This can be done by implanting inside the optical cavity a nonlinear optical material, in which photons can interact through intensity-dependent, so-called $\chi^{(3)}$ contributions to the index of refraction. Those contributions are ordinarily too weak to allow photons to thermalize during the cavity lifetime, but one can enhance them by tuning the photons' energy to that of an electronic excitation in the medium. The excitation is typically an exciton—a bound electron–hole pair—in a semiconductor quantum well, but any electronic excitation with a well-defined transition energy will do. The excitation gives rise to a resonance in the polarizability of the medium, which sharply increases the nonlinearity of the index of refraction and strengthens the photon–photon interaction. An alternative approach to thermalizing photons is to choose a material whose absorption and emission spectra are related by a Boltzmann factor and operate the optical cavity in a regime where photons are repeatedly absorbed and reemitted before escaping.[4]

When the coupling between a photon and an exciton becomes strong enough, one can no longer think of the two particles as separate eigenstates; instead, one must talk of a polariton—a quantum superposition of a photon and the electronic excitation.[2,3] The resulting dispersion relation, shown in figure 2, is more complicated than that of an isolated photon; it exhibits an upper branch and a lower branch, each of which is a mixture of the original photon and exciton states.

Polaritons in the lab

In a way, detecting polaritons in experiments is as simple as detecting light. Although polaritons are not pure photons, they couple directly to photons outside the host optical cavity, and they can therefore be created by laser excitation and observed by photon detection. In a typical experiment, a weak pump source generates polaritons, and the polaritons' momentum and energy distributions are inferred from the angular distribution and spectra of the cavity emission. The pump source is typically tuned to an energy far away from the ground state of the polaritons in the cavity. Polaritons can also be pumped electrically, but it's challenging to inject electrons in a way that preserves the quality of the cavity and the lifetimes of the polaritons.

More than a decade has now passed since the first experiments clearly demonstrated polariton condensates.[5,6] In those experiments, condensates were evidenced by the emergence of a sharp peak near the ground state in the momentum distribution as shown in figure 3a, a jump in the spatial and temporal coherence of the emitted light, and the spatial condensation of polaritons in a trap.

Like most subsequent experiments, those early studies used either gallium arsenide or cadmium telluride quantum wells as the active layer. Unlike silicon, those semiconductors have a direct bandgap—that is, photons can directly excite electrons from the top of the valence band to the bottom of the conduction band. Thus the lowest-energy excitons are optically active and can easily couple to cavity photons to form polaritons. Extensive R&D on GaAs and CdTe have perfected techniques for growing those materials into high-quality, low-structural-disorder devices. The two materials are therefore well suited to the accurate fabrication of complex heterostructures.

The quantum wells are usually sandwiched between two distributed Bragg reflectors, which act as the two cavity mirrors. Distributed Bragg reflectors consist of alternating layers of semiconductors having differing refractive indices. If each layer is a quarter of a wavelength thick, then the reflections from each interface interfere constructively. As the number of layers increases, the total reflectivity approaches unity.

In early condensation experiments, leakage through the

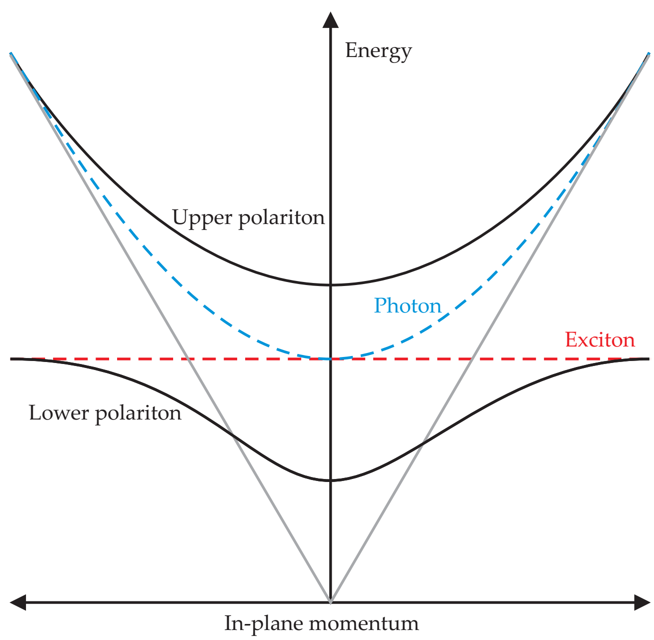

FIGURE 2. THE DISPERSION RELATION FOR A PHOTON in an optical cavity is parabolic for small in-plane momentum and converges to the free photon dispersion (gray lines) at large momentum. When one mode is tuned to the energy of an electronic excitation, or exciton, photon–exciton coupling inside the cavity gives rise to new hybrid modes (black), known as polaritons.

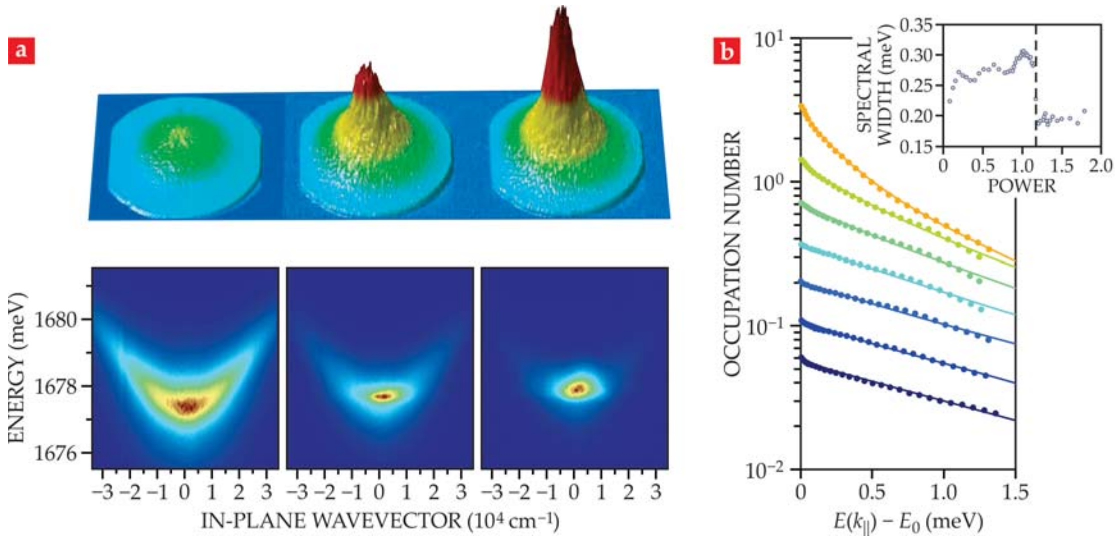

FIGURE 3. THE BOSE–EINSTEIN CONDENSATION OF POLARITONS carries clear, experimentally observable hallmarks. **(a)** As polariton density increases (left to right) above the condensation threshold, the polaritons' in-plane momenta, evidenced by the angular distribution of their light emission, peaks sharply at zero (top), and their energies and momenta converge on the minimum of the dispersion curve (bottom). That curve shifts slightly upward due to interaction effects as the polariton density increases. Here, regions of parameter space colored red correspond to those that produced the highest density of photon counts. (Adapted from ref. 5.) **(b)** In experiments that achieve full equilibrium, the evolution of the observed energy distribution (dots), plotted as a function of wave number $k_\parallel$, is well described by the Bose–Einstein distribution (curves). Here, polariton density increases as curves change from purple to orange, with the orange curve corresponding to a system on the verge of condensation. The inset shows the sharp decrease in the spectral width of the polariton emission line as pump power crosses the threshold for condensation, indicated by the dashed line. (Adapted from ref. 7.)

mirrors prevented the polaritons from fully equilibrating, and number conservation was only approximate. More recently, improved Bragg reflectors have yielded photon lifetimes long enough to achieve equilibrium condensation of polaritons.[7] (See figure 3b.) The latest experiments have also produced a wide range of condensation-related phenomena beyond those seen in early demonstrations, including Josephson oscillations of two coupled condensates, phase locking of two or more condensates, quantized vortices, and the superfluid-like suppression of scattering from defects. For a general review of those and other effects, see the chapters on polariton condensation in reference 8.

Lasing, condensation, and superfluidity

Because a hallmark of polariton condensation is the emission of coherent light, it's natural to ask how polariton condensates differ from lasers. The most notable distinction is that a polariton condensate ideally exists in thermal equilibrium, its various modes occupied according to the Bose–Einstein distribution. The laser, by contrast, exists in a highly nonequilibrium state, in which the occupation of modes is set by the balance of gain and cavity loss. The need to produce gain means that lasing typically requires inversion: The gain medium must be sufficiently excited that the rate of emission exceeds that of absorption.

In general, using the language of emission and absorption implies that the system has incoherent transfers of excitation between matter and light. This in turn implies weak coupling between the photons and the medium. In the strongly coupled, polariton limit, the eigenstates are coherent superpositions of matter and light excitations. In that limit, the optical medium exhibits not stimulated photon emission but stimulated polariton–polariton scattering, whereby the bosonic particles scatter preferentially into states that are highly occupied, due to the $(1 + n)$ final state factor that comes from quantum statistics. Stimulated scattering between different modes leads to macroscopic occupation of the ground state—that is, to condensation.

Because the condensate's coherent emission does not require stimulated emission, neither does it require inversion. Interestingly, both polariton condensation and standard lasing can be achieved with the same device; standard lasing occurs in the regime of weak light–matter coupling, at pump intensities two or three orders of magnitude larger than the threshold for polariton condensation.[9]

Is a polariton condensate a superfluid? In the limit where the polariton lifetime is much longer than the thermalization time, the answer should be yes. Any gas of weakly interacting bosons becomes a superfluid at low enough temperatures. However, the polariton's finite cavity lifetime complicates the picture. Theory confirms that standard effects of superfluidity can survive over small length scales in polariton condensates but that the asymptotic behavior at large length scales is not superfluid. (That asymptotic result applies only to two-dimensional condensates, but because polariton condensates are typically formed between planar mirrors, they are naturally two-dimensional.) The theory raises interesting questions about the fundamental differences between the phases of bosons with finite lifetimes and the phases of those that live indefinitely. See chapter 11 of reference 8 for a review of the key issues. Groups experimenting with polariton condensates have seen superfluid-like phase correlations and scattering suppression[10] but have yet to directly measure the superfluid density of a polariton condensate.

An optics playground

To appreciate polaritons' technological promise, it is helpful to think of them from a nonlinear optics perspective—that is, as strongly dressed photons. In that view, polaritons' chief advantages are their strong nonlinearity relative to typical photonic systems, their ability to thermalize, and their ability to emit laser-like beams under low power.

A long-standing goal of optical communications research is to make an all-optical transistor, in which one light beam, the

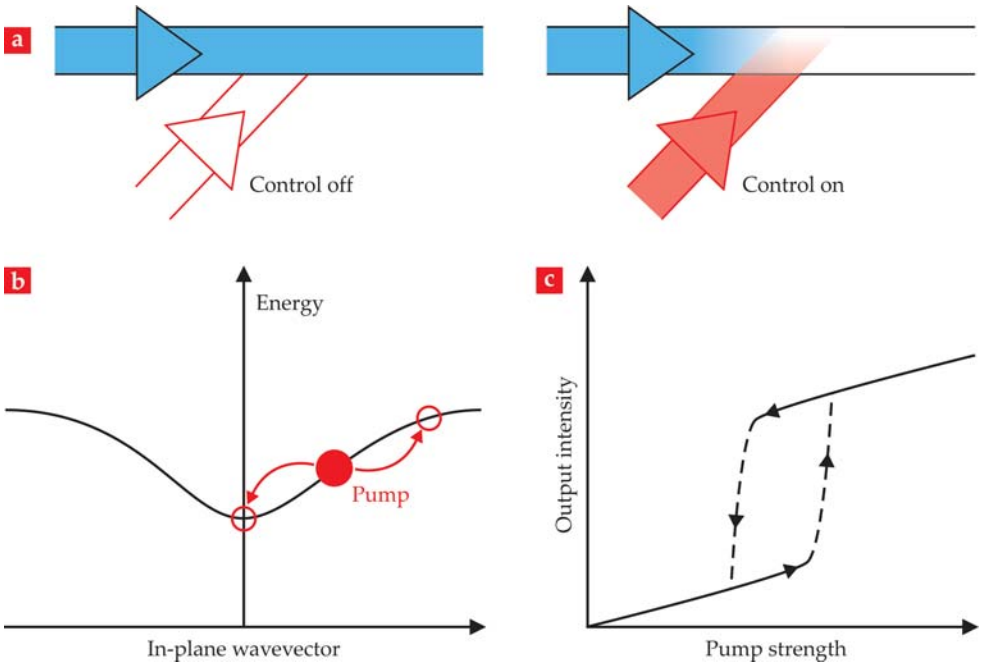

FIGURE 4. OPTICAL SWITCHES can be achieved through various polariton-based schemes. **(a)** An optical transistor design exploits repulsive interactions between polaritons: The flow of source polaritons (blue) from left to right shuts off when a gate beam (red) is turned on and resumes when it is turned off. **(b)** Source polaritons injected into a cavity by a pump beam at an energy and momentum near the inflection point of the polariton dispersion curve can be stimulated by gate polaritons to scatter into low- and high-momentum states. **(c)** Due to a nonlinear relationship between polaritons' energy and density, bistability of the output intensity can be obtained by using an appropriately tuned pump beam.

gate, directly modulates the intensity of another light beam, the source, without the need for electronics.[11] Because polariton systems are highly nonlinear, they can be hypersensitive to external inputs and can potentially serve as the optical switches underlying such transistors—possibly in integrated polaritonic circuits. Several schemes have been proposed to that end.[12]

In one approach, illustrated in figure 4a, the repulsive interaction between polaritons is harnessed to spatially steer light: Polaritons created by a gate beam can repel and redirect a source beam of moving polaritons along a desired path.

Another transistor scheme exploits stimulated scattering: The gate beam creates a dense region of polaritons in one state, and source polaritons passing through the system preferentially jump into that state. In the example illustrated in figure 4b, a strong pump beam—the source—is injected at the energy and momentum near the point of inflection on the polariton dispersion. The resulting polaritons are modulated by the injection of a gate beam that seeds the optical cavity with a small population of condensed polaritons in the $k_\parallel = 0$ mode. Pairs of source polaritons are stimulated to scatter into the $k_\parallel = 0$ mode and into a mode at twice the pump momentum. Such a device can work as an optical amplifier, and early demonstrations with GaAs-based cavities have shown record optical gains approaching a factor of 100.

A third approach to optical switching exploits the dependence of the polariton dispersion on particle density. Interactions between the polaritons cause the particles' energy to grow as their density increases. As a result, the cavity's resonance frequency shifts, which can cause its absorption and reflection properties to change dramatically. As illustrated in figure 4c, the effect can produce bistability. If a pump laser is tuned to slightly higher than the polariton resonance frequency, it generates a positive feedback effect: As light is absorbed and the population of polaritons grows, the polariton resonance shifts upward, toward the pump frequency. That feedback enhances the absorption, which further increases the polariton population and resonance. The effect stabilizes when the resonance frequency reaches the pump laser frequency. Then, even if the pump intensity subsequently drops, the absorption remains strong.

The ability of polaritons to thermalize and condense into a superfluid could also lead to novel applications. For instance, superfluid polariton condensates might allow optical signals to flow through circuits without dissipation. Also, unlike a conventional optical waveguide, a polariton-based circuit would be able to implement frequency-shifting schemes: Stimulated scattering allows a polariton condensate to quickly shift to the lowest energy state of a system; the excess energy is dumped to the surrounding solid. Thus, for example, an input beam with fluctuating frequency, or multiple input beams with different frequencies, could be used to modulate an output having a single, stable frequency.

Because polariton condensates produce coherent light without the need for the inversion of a gain medium, they can potentially serve as ultralow-threshold coherent light sources. Such devices could be useful in situations that call for low power consumption or low heat production, such as in densely packed integrated circuits. They may also allow coherent emission in materials that otherwise wouldn't lase due to nonradiative losses at high current densities.

To serve as viable laser alternatives, polariton devices would need to operate in a regime quite different from that used to study condensation. To demonstrate that a condensate has thermalized, for instance, one wants a cavity geometry that sustains many polariton modes so that the occupation of those modes can be plotted as a function of wavevector. In contrast, lasing applications benefit from geometries that are explicitly optimized to control and select for a single mode. Such single-mode polariton devices have successfully demonstrated high coherence with low power thresholds.

Polaritons may also provide important sources in the hard-to-access yet technologically useful terahertz band of the electromagnetic spectrum. Experiments have shown that stimulated transitions between polariton states in noncentrosymmetric devices can be engineered to produce terahertz emissions.[13] (In centrosymmetric materials, selection rules prevent the emission of electromagnetic radiation via transitions between polariton states.) Such sources would have potential applications in biosensing, security screening, and other areas.

Some researchers also propose that polaritons can mediate superconductivity.[14] The mechanism closely parallels that of exciton-mediated superconductivity, proposed half a century ago by Vitaly Ginzburg; the exciton interactions facilitate the Cooper pairing of electrons in much the same way that phonons do in traditional superconductors. Although no studies to date have produced polariton-mediated superconductivity, there have been promising developments with 2D materials, in which researchers have demonstrated the ingredients required to test such an idea: strong matter–light coupling in gate-doped layers of electrons.[12,15]

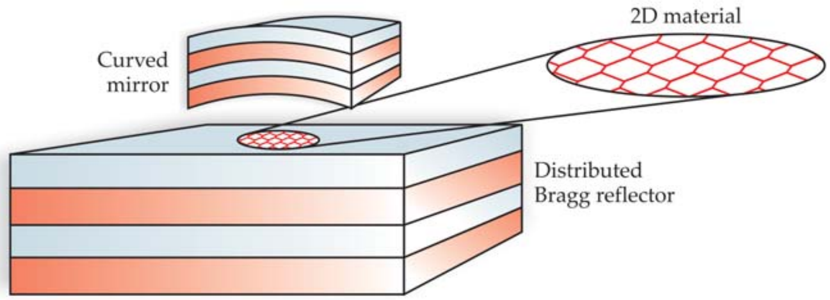

FIGURE 5. IN AN OPEN-CAVITY DESIGN, the active layer rests atop a bottom mirror. A separate top mirror—usually curved to better focus light—can scan freely in the lateral direction. Open cavities are commonly used to create polaritons in two-dimensional flakes of transition metal dichalcogenides, organic materials, or other active materials that can't easily be integrated into sandwiched heterostructures.

A materials medley

In principle, there is no reason polariton condensates cannot form at room temperature. The cavity size and the active layer's refractive index can be adjusted to make the effective polariton mass as light—and the critical temperature as high—as one likes. The problem is that the polaritons themselves can't take the heat: Warm temperatures destroy the strong matter–light coupling and cause excitons to thermally dissociate into electrons and holes. In GaAs and CdTe heterostructures, for instance, condensates must be formed at cryogenic temperatures, typically in the range of 4–70 K. That's high compared with cold-atom condensates but still too cool to be practical for most real-world applications.

A room-temperature condensate would require a material with an exciton binding energy much greater than $k_B T$, equal to 25 meV at room temperature. The matter–light coupling must be such that the energy splitting between the upper and lower polaritons also far exceeds $k_B T$. That splitting depends both on the oscillator strength—a unitless parameter that indicates the strength of the response of the electronic transition to light—and on the emitter density. Three main classes of materials are being pursued with an eye toward room-temperature polariton condensation: wide-bandgap semiconductors, 2D materials, and organic materials.

The first class, wide-bandgap semiconductors, includes materials such as gallium nitride and zinc oxide, which operate on the same principles as GaAs and CdTe but have exciton binding energies exceeding 50 meV, easily high enough for excitons to persist at room temperature. Methods for growing and fabricating devices with those materials are relatively immature, so combining the materials with high-quality mirrors and reducing the effects of disorder remain challenges.

Two-dimensional materials are interesting candidates because, in contrast to 3D semiconductors, they have no out-of-plane charges to screen the Coulomb interaction between an electron and its hole. That yields large exciton binding energies of up to 500 meV or more. Those more tightly bound excitons exhibit a smaller Bohr radius—that is, less separation between the electron and hole—and therefore increased electron–hole overlap. Because the matter–light coupling is proportional to the probability that the electron and hole meet each other, the coupling strength of the exciton resonance increases too.

Unlike experiments with standard semiconductors, experiments with 2D materials are often performed in open optical cavities, with the top and bottom mirrors grown on separate substrates, and often with flakes of the active excitonic 2D material strewn on the bottom mirror. The top mirror is typically grown on a curved surface so that it can be scanned and focused on individual flakes, as illustrated in figure 5.

So far, efforts to develop 2D materials have centered on transition-metal dichalcogenides, such as molybdenum diselenide. Preliminary experiments[15] with single-layer flakes of $MoSe_2$ inside an optical microcavity have produced a polariton splitting of 20 meV.

The third class of candidates, organic materials—including ones made up of small molecules or conjugated polymers—opens vast new possibilities for polariton condensates. Organic materials generally support Frenkel excitons, delocalized electronic excitations of single molecules.[16] Their binding energies are on the order of 1 eV, and they can easily persist at room temperature. In addition, the dipole moments for Frenkel exciton transitions are large enough to give rise to polariton splittings ranging from 100 meV to 1 eV or more.[17,18] The largest couplings approach the ultrastrong coupling limit, wherein the matter–light coupling strength is comparable to the bare optical transition frequency. The formation of polaritons can thus change the molecule's electronic ground state.

Organic molecules present their own challenges, however. Even in an isolated organic molecule, the photophysics can be complex. Strong coupling of light to vibrational modes can lead to large Stokes shifts between absorption and emission lines. In disordered, long-chain polymers, different chromophores may have different energies, and those energies may depend on the environment and the polymer conformation, all of which complicates energy-transport and relaxation dynamics. Those issues are being actively studied not only to improve organic-based polaritonic devices but to explore the effects of matter–light coupling on photophysical processes in organic photovoltaics and light-emitting devices.

Although organic materials are relatively easy to fabricate using solution processing, spin coating, and other methods, their fragility limits the ways they can be combined with other materials. The high temperatures and sputtering materials used to create distributed Bragg reflectors, for instance, can damage organic materials. Here again, open-cavity structures can be highly effective because they allow one to avoid depositing a second mirror onto the fragile active layer.

Some materials science issues remain to be addressed for organic materials to realize their full promise. Applications such as polariton integrated circuits that require polaritons to travel macroscopic distances call for longer polariton lifetimes and lower disorder than have been achieved with organic semiconductors. Also crucial is the strength of polariton–polariton interactions, particularly in applications involving polariton switches and transistors. Interactions between Frenkel excitons tend to be suppressed, and although some hallmarks of polariton condensation have been seen in organic materials, others—including parametric scattering and optical amplification—remain elusive.

Just warming up

Over the past decade, researchers have developed a host of strategies for putting polariton condensates to use—at ever higher temperatures and in increasingly diverse materials. Many potential applications have already been demonstrated at low temperature, and the list of promising materials for room-temperature condensation is also growing. Efforts to develop polariton applications may beget new materials and structures, each tailored and optimized for specific uses. Hybrid devices that combine the advantages of different materials may also provide a way to optimize polariton properties for particular applications.

Perhaps no development would be more transformative than fabrication of a polariton-based transistor. Despite decades of optics research, no practical optical transistor yet exists: Current versions are too big to be used in integrated circuits, require very high light intensity, suffer high losses, or have poor on–off ratios. It is not a stretch to say that an efficient optical transistor will someday radically change the nature of optical communications much the way the electronic transistor once changed electronics. Neither is it a stretch to say that that day may be nearly upon us.

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