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REVIEW

Spontaneous Bose Coherence of Excitons and Polaritons

David Snoke

In the past decade, there has been an increasing number of experiments on spontaneous Bose coherence of excitons and polaritons. Four major areas of research are reviewed here: three-dimensional excitons in the bulk semiconductor Cu_2O, two-dimensional excitons in coupled quantum wells, Coulomb drag experiments in coupled two-dimensional electron gases, and polaritons in semiconductor microcavities. The unifying theory of all these experiments is the effect of spontaneous symmetry breaking in the Bose-Einstein condensation phase transition.

Numerous recent experiments have studied coherence in semiconductor systems. In most of these experiments, the coherence is generated from a coherent laser that excites the system. The laser itself acquires coherence because a selected optical state is amplified. In the past two decades, however, several experimenters have pursued the possibility of spontaneous coherence in semiconductor systems. In such a case, no single state is selected for amplification. Instead, coherence appears in a state selected by the system itself by means of a thermodynamic phase transition.

The phase transition that can cause this is Bose-Einstein condensation (BEC) of excitons or polaritons. In this phase transition, a macroscopic number of particles enter a single quantum state, forming a coherent state with definite phase by the process known as "spontaneous symmetry breaking" (*1*). The underlying physics is the same as BEC of atoms, seen in superfluid helium or, more recently, in alkali atoms in magneto-optical traps (*2*, *3*) and in spin-polarized hydrogen (*4*). Excitons and polaritons, quanta of excitation that are integer-spin bosons, often have an effective mass and a long lifetime, so that they can be treated theoretically as metastable atom-like particles. As such, the laws of thermodynamics apply and BEC is expected at a low temperature, typically a few kelvin. After initial proposals in the 1960s (*5*, *6*), the theoretical basis for Bose condensation of excitons was laid down in a number of papers (*7*–*10*) in the 1970s and 1980s and has continued to attract theoretical interest (*11*–*13*).

Department of Physics, University of Pittsburgh, 3941 O'Hara Street, Pittsburgh, PA 15260, USA.

Phase Transitions of Excitons

As excitations of the medium, excitons and polaritons are not strictly distinguishable from photon states. Figure 1 shows a typical dispersion relation in a bulk semiconductor. At energy well below the band gap, the excitations are photon-like and have the dispersion relation

$$\hbar\omega = \hbar\left(\frac{c}{n}\right) k \qquad (1)$$

where $\hbar$ is Planck's constant divided by 2π, ω is the photon frequency, c is the speed of light, n is the index of refraction of the medium, and k is the photon wave number. In the opposite limit, at high energy and momentum, one can view the fundamental excitation of the medium as taking an electron

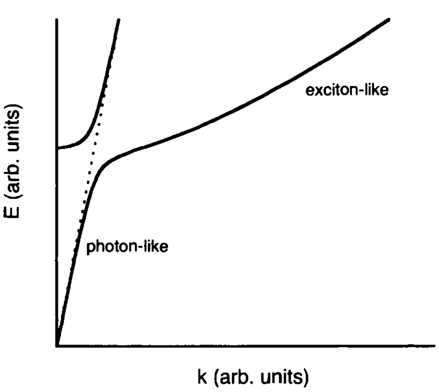

Fig. 1. A typical polariton dispersion relation in a semiconductor. The exciton energy E, alone, is given by Eq. 2. At low momentum, the exciton state mixes with the photon states in the medium. These mixed states are called polaritons. The dashed line gives the photon dispersion without mixing.

from the valence band to the conduction band, leaving behind a hole (an empty state) in the valence band. In typical semiconductors, both the electron in the conduction band and the hole in the valence band move freely, with an effective mass on the order of the free electron mass in a vacuum. The Coulomb attraction between the negative electron and the positive hole leads to the bound states known as Wannier excitons. The energy of the excitations in this limit is simply given by the band gap energy (E_{gap}) minus the Rydberg binding energy of the excitons, calculated using the hydrogenic Rydberg formula, plus the kinetic energy of the pair

$$\hbar\omega = E_{gap} - \frac{e^4 m_r}{2\epsilon^2\hbar^2} + \frac{\hbar^2 k^2}{2m_{tot}} \qquad (2)$$

where e is the electron charge, ε is the dielectric constant of the medium, m_r is the reduced mass, and m_{tot} is the total effective mass of the electron and hole. When the photon energy is comparable to the exciton energy, the curves given by Eq. 1 and Eq. 2 cross, and mixing of the states occurs (Fig. 1). The excitations in the region of the mixing are known as polaritons. As seen in Fig. 1, the photon states merge continuously into the polariton states, which merge continuously into the exciton states. All three of these states are bosonic states; that is, they obey the law of stimulated emission derived from the properties of the bosonic quantum field operators.

Some researchers over the years have preferred to say that excitons and polaritons are only approximately bosons, because at high density the Fermi statistics of the underlying electrons and holes become important. Of course, atoms such as helium and hydrogen are only approximately bosons in this sense, also, as they are constructed from fermions, and the Pauli exclusion from these component particles becomes significant at high density. The relevant issue, however, is whether the interactions between the particles are strong enough to destroy the pair correlations of the fermion components, so that the system is characterized only by Fermi statistics. This occurs, for example, at high temperature and high density when excitons become ionized into electron-hole plasma (*14*), and in electron-hole liquid (EHL), also known as electron-hole droplets, a liquid phase analogous to liquid mercury, extensively studied in the 1970s and 1980s in bulk indirect-gap semiconductors (*15*). In many semiconductor systems, however, EHL is forbidden. At low density and low temperature in these systems, the effect of Pauli exclusion can be accounted for simply as a hard-core repulsion, and the excitons should behave as a weakly interacting Bose gas (*8*, *11*), so that BEC becomes possible. At low enough temperature, spontaneous Bose coherence can occur even at high density. As shown in early work (*7*, *10*), the theory of the electron-hole pairs at zero temperature allows them to transform continuously from a BEC into a superfluid state analogous to Bardeen-Cooper-Schreiffer (BCS) superconductivity as density is increased.

A more important issue is the relatively short lifetime of excitons and polaritons as compared to that of atoms. Excitons and polaritons normally convert into photons, which leave the system. For a condensate to appear spontaneously according to the laws of thermodynamics, the lifetime of the particles in the system must be long compared to their thermalization time; that is, the time needed for the system to have a definable temperature. In this case, the particles are in quasi-equilibrium, and equilibrium thermodynamics can be applied. For many exciton or polariton systems, however, the lifetime and the thermalization time are comparable, on the order of picoseconds. This problem can be overcome by choosing or designing systems with long exciton or polariton lifetime.

In the 1980s, there was some question about exactly how long the lifetime must be to have a true quasi-equilibrium. Both the experiments on atomic BEC and theoretical models (*16*) have shown that the time scale for the onset of thermodynamic BEC is on the order of a few classical scattering times. If the exciton lifetime is long compared to this (tens to hundreds of nanoseconds as compared to tens of picoseconds for thermalization by exciton-phonon interaction) then BEC should be possible. At high exciton density, the thermalization time can be even shorter because of exciton-exciton interactions.

Experiments with Excitons in Cuprous Oxide

One way to produce excitons with a very long lifetime is to use a semiconductor in which radiative recombination from the lowest exciton state, and therefore also the polariton

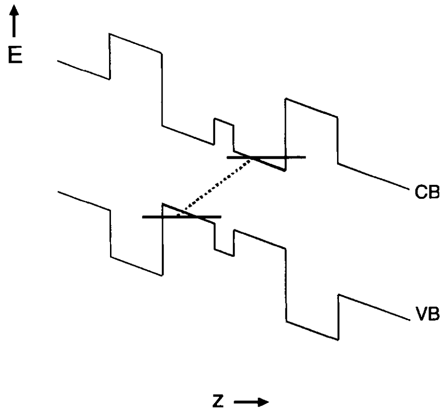

Fig. 2. The energy bands of coupled quantum wells, in the direction perpendicular to the planes in a layered heterostructure. When an electric field is applied, the bands tilt, which causes the electrons and holes to be spatially separated into two different wells. This leads to longer exciton lifetimes, because the electrons and holes must tunnel through a barrier to recombine with each other, and also to a decrease in the gap energy, known as the quantum confined Stark effect. CB, conduction band; VB, valence band; z, distance in the direction perpendicular to the planes.

effect, is forbidden by symmetry. Cu_2O is an example of such a crystal; the lifetime of the excitons can be tens of microseconds at low temperature (*17*), and lifetimes of hundreds of nanoseconds are routinely measured.

Several studies in the early 1990s reported evidence for Bose condensation of excitons in Cu_2O. In one set of experiments (*18*), the spectral line shape of the lumiscence was analyzed for evidence of Bose condensation. At low exciton density, this procedure is valid because the phonon-assisted luminescence from Cu_2O gives the instantaneous energy distribution function of the excitons at all times (*19*). In quasi-equilibrium, the luminescence spectrum of Bose-condensed excitons should have a distinctive peak at low energy (*20*). At high exciton density, however, the exciton lifetime in Cu_2O is shortened by an Auger nonradiative process, in which two excitons collide and one is annihilated, giving its energy to ionizing the other. This process not only shortens the exciton lifetime, it also heats the exciton gas. Recent studies (*21*) have shown that the lifetime at the highest densities decreases to become comparable to the thermalization time of the excitons, so that it is no longer valid to assume that the luminescence line shape of the excitons indicates a quasi-equilibrium distribution.

Another set of studies on Cu_2O (*22*), performed at very low exciton density at which Auger recombination should be negligible, has shown ballistic transport of a packet of excitons at the speed of sound in the crystal below a critical temperature, as well as amplification of the exciton pulse when other excitons are added. Because another property of a BEC is superfluidity, the ballistic transport of the excitons was taken as evidence of BEC. Theoretical analysis (*23*) has indicated that this effect may simply be related to the high purity of the samples of Cu_2O used, which allows very high diffusivity of the excitons at low temperatures, in combination with a "phonon wind": a drift force arising from hot nonequilibrium phonons created by the same laser that creates the excitons.

A general problem with Cu_2O is that because the luminescence from the lowest exciton state (the singlet "paraexciton") is forbidden, it is difficult to observe them. For this reason, many studies have concentrated on the higher-lying, triplet "orthoexciton" state, for which luminescence is weakly allowed. If the lifetime for conversion by spin flip to paraexcitons is slow, a quasiequilibrium state can in principle be reached among the orthoexcitons.

One set of experiments on Cu_2O created orthoexcitons directly in their lowest energy state using a laser tuned to their ground-state energy (*24*). A sharp persistent peak was seen in the phonon-assisted luminescence. In this case, however, it is debatable whether to call this state a BEC, because the coherence of the pump laser was coupled directly to the exciton states, so that there was apparently no spontaneous exciton coherence. The same has been argued of similar experiments using two-photon resonant creation of orthoexcitons in Cu_2O (*25*) and of biexcitons (bound states of two electrons and two holes) in the semiconductor CuCl (*26*, *27*). In these cases, one can talk of a "driven BEC," because a Bose condensate is interchangeable with a classical coherent wave (*1*). Some object to this terminology, however, because the term condensation implies a thermodynamic phase transition to many people.

One of the appeals of experiments in Cu_2O is that the excitons can be held in a harmonic potential trap exactly analogous to the magneto-optical traps used for alkali gases. A method of trapping the excitons in a harmonic potential inside the crystal using stress has long been known (*28*). The applied stress, however, causes two effects that shorten the exciton lifetime: decay of the paraexcitons into photons becomes allowed, and the Auger process appears to become stronger (*29*). It may be possible, however, to create a harmonic potential trap with very weak stress, so that the exciton lifetime is not strongly altered.

Recent work has been encouraging for experiments on Bose condensation of excitons in Cu_2O. Although emission from paraexcitons is very weak at zero or near-zero stress, they can be detected by differential absorption of a laser tuned to a higher exciton state (*30*). The possibility still remains, therefore, for observation of a BEC state of paraexcitons at very low temperature and low excitation density.

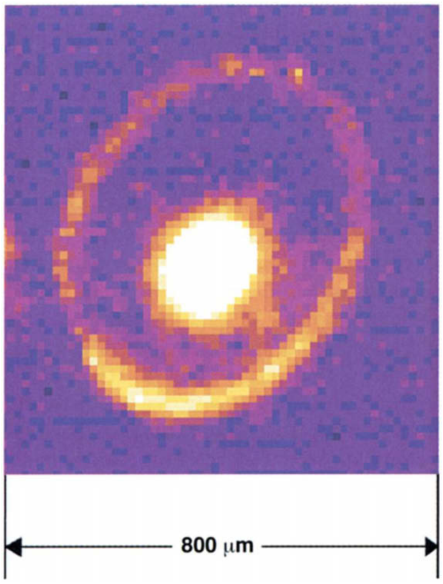

Fig. 3. A false-color image of the ring of exciton luminescence that appears in recent experiments on excitons in coupled quantum wells. The excitons are created in the central laser spot, which has bright luminescence at all densities. Under certain conditions, they travel long distances without emitting luminescence. This may be evidence of exciton superfluidity. This image is taken from the same experiments as those in (*45*).

Indirect Excitons in Coupled Quantum Wells

Another way to produce long exciton lifetimes is to use the band-gap engineering of quantum heterostructures. Figure 2 shows a typical coupled quantum well structure. When an electric field is applied perpendicular to the wells, the electrons and holes are pulled into adjacent wells. If the barrier between the two wells is thin but high, the electrons and holes will feel the Coulomb attraction of each other, forming excitons, but the wave functions of the electrons and holes will have very little overlap, so that the excitons have a long lifetime. Lifetimes of hundreds of nanoseconds have been recorded in typical structures (*31*).

In two dimensions, true BEC is not strictly possible, but a related phase transition known as the Kosterlitz-Thouless superfluid (KTS) transition can occur (*32*). In this case, spontaneous phase coherence of the particles occurs but does not persist to infinite range. The physical basis for BEC and the KTS phase transitions is the same: The bosonic particles prefer to be in the same quantum state, which leads to spontaneous phase correlation. Several theoretical works have treated the topic of KTS of excitons in two dimensions (*13*, *33–35*).

An early study on this type of structure (*36*) reported evidence of Bose condensation based on luminescence line shape analysis. The line shape of the luminescence was later reinterpreted (*37*) to arise from excitons trapped in local minima caused by inhomogeneities in the structure. This points out an intrinsic difficulty in the study of KTS of excitons in heterostructures. The critical temperature for this phase transition is proportional to the exciton density (*32*). Because the excitons must have low density in order to avoid Fermi phase-space filling and screening effects, the excitons must be kept at low temperature. At low temperature, however, the excitons tend to become trapped in local minima, because the technology of fabricating these structures tends to include defects of various types.

This barrier may have finally been overcome in nearly perfect heterostructures fabricated in recent years. Several experimental groups have reported evidence for KTS in this type of structure, including increased diffusivity (*38*), slow fluctuations of the luminescence (*39–41*), spectral narrowing (*42*), and enhanced scattering (*43*). Recently, a striking effect of long-range transport of excitons in nonluminescing states, known as dark states, has been seen (*44*, *45*). This effect is observed in a large ring of luminescence from the excitons (Fig. 3), with a radius of hundreds of microns up to millimeters. The ring appears only above a critical density, which increases with increasing temperature.

The transport of the exciton signal over such a long distance without luminescence is not well understood. Normally, in thermal equilibrium, excitons in quantum wells emit light at all times. One possibility is that carriers are trapped in metastable high-energy states, although such an effect has not been observed before. Another possibility is that excitons in a superfluid state do not emit light because of some special property of the condensate. In this model, the superfluid fills the region between the laser excitation spot and the ring, and the luminescence occurs only at the boundary of the superfluid region, when the exciton density drops below the critical density and the excitons revert to the normal state.

These experiments give only very indirect evidence for a KTS state of spontaneous coherence, because no direct measurement is made of the dark states occupied by the excitons. To truly show spontaneous phase coherence, one would need a measurement of the coherence of the exciton state such as an interference measurement. The macroscopic motion of the excitons is nevertheless fascinating. Earlier studies (*46*, *47*) have shown that stress and electric field can be used to apply a force on the excitons in these structures, even though they are charge-neutral. One can therefore imagine circuits in which excitons are moved from one place to another in a heterostructure over millimeter distances.

Exciton Condensate in Coulomb Drag Experiments: Experiments with Permanent Excitons

A similar system of coupled quantum wells has been developed that can be viewed as having permanent excitons with infinite lifetime. In these experiments, two parallel quantum wells are created separated by a thin barrier, as in the double quantum well experiments discussed above. The two wells are given a permanent population of electrons, however, by doping in a structure similar to a field-effect transistor. When a strong magnetic field is applied normal to the wells, the electrons are confined to Landau orbitals, which have a total density of states that depends on the value of the magnetic field. If the magnetic field is tuned to the right level, both quantum wells will be exactly half filled. In this case, one can view each of them as having an equal number of electrons and holes. The Coulomb attraction between electrons and holes in the adjacent quantum wells causes them to pair into excitons (Fig. 4). Because of the relatively high density of electrons, with spacing between the particles comparable to the exciton Bohr radius, the excitons cannot be viewed as weakly interacting bosons, but at low temperature they form a BCS-like state (*48*, *49*). Because this only occurs at very low temperature, these experiments are done at millikelvin temperatures in a dilution refrigerator.

Because the excitons never convert into photons, their state can only be probed by a transport measurement. This is done by means of Coulomb drag, in which a current is induced in one of the layers, and the resulting voltage is measured in the adjacent layer. When the electrons go into the excitonic state, the holes will be dragged along with the electrons, resulting in a much higher Coulomb drag voltage (*50*, *51*).

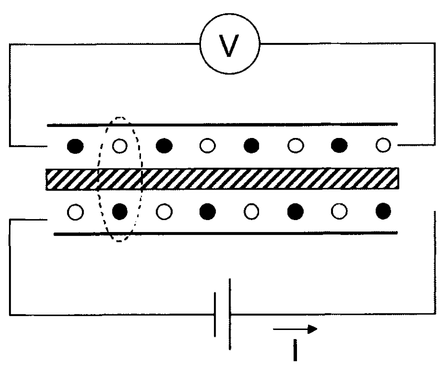

Fig. 4. Idealized picture of a Coulomb drag experiment on excitons in coupled half-filled Landau levels. When each level is half filled, it can be viewed as having equal numbers of electrons and holes. Electrons in one layer can form exciton-like pairs with holes in the other layer. The Coulomb attraction between these pairs causes the charge in one layer to tend to follow the charge in the other layer. *V*, voltage; *I*, current.

This system has some similarity to early proposals (*52*, *53*) for a permanent excitonic condensate in double-well narrow-gap semiconductors. If a semiconductor system can be constructed that has exciton binding energy greater than the band gap between the valence and conduction bands, a macroscopic instability will occur in which electrons at the top of the valence band can lose energy by forming excitons with holes in the conduction band. This state, called the excitonic insulator or excitonium, should transform a narrow-gap semiconductor into an insulator. The paired state of the carriers is not expected to be a phase-coherent state, however (*54*, *55*), but corresponds to a real-spacing ordering that should have observable macroscopic properties. Experimental evidence for the excitonic insulator state, consisting of a peak in the resistance of the material as the band gap is varied, has been reported for both three-dimensional (*56*) and two-dimensional (*57*) systems. It has also been suggested that narrow-gap Kondo insulators have this character (*58*).

Microcavity Polaritons

In the past decade, there has been extensive work on Bose effects of polaritons in microcavities. A microcavity simply consists of two mirrors opposite each other, as in a laser cavity, which are built into a heterostructure (Fig. 5A). The two mirrors are typically fabricated as sets of dielectric layers forming a Bragg reflector.

If the Bragg reflectors have spacing L, the photon modes between the two mirrors have the dispersion relation

$$\hbar\omega = \hbar\left(\frac{c}{n}\right)\sqrt{k^2 + \left(\frac{2\pi}{L}\right)^2} \quad (3)$$

where k is the in-plane momentum. If the spacing of the layers is chosen properly, the wavelength of light reflected by the mirrors can be nearly exactly the same as that of the exciton in a quantum well between the mirrors. This leads to a mixing of the two states, which gives the dispersion relation shown in Fig. 5B. The lower polariton branch has a minimum at $k = 0$ with a very small effective mass, so that these particles act like atoms in two dimensions and therefore can in principle undergo a KTS transition. Because the critical temperature for the KTS transition varies inversely with the particle mass (*32*), the light effective mass allows a KTS transition at lower particle density than that of typical excitonic condensates, helping to avoid the high-density effects discussed above.

When a photon is absorbed in the microcavity structure, the in-plane component of the photon momentum is conserved, coupling directly to the polariton mode with the same in-plane momentum. This means that a specified polariton state can be pumped by choosing the angle of incidence of the laser beam that creates the polaritons. In the same way, the number of polaritons in a specific state can be detected by observing the photon emission only at a selected angle. Because the angle of incidence can be determined very accurately, these experiments therefore offer the possibility of studying the polariton momentum distribution near $k = 0$ with very high accuracy.

The confinement in the mirrors has the effect of decreasing the exciton lifetime, because emission is enhanced in the direction perpendicular to the mirrors. Typical lifetimes of polaritons in these structures are on the order of picoseconds, so that it is not clear whether a thermodynamic Bose condensation can occur. Several studies (*59–61*) have aimed at demonstrating the nonequilibrium effect of stimulated scattering, in which scattering into a final state is enhanced by the bosonic factor $(1 + n_f)$, where n_f is the number of particles in the final state. This final state term is the same for all bosons, whether photons or atoms, and is responsible both for stimulated emission of photons and for the buildup of BEC (*16*, *19*). In a typical experiment, a polariton state with $k \neq 0$ is pumped, and polariton-polariton elastic scattering of the population in this state leads to two additional populations, which correspond to $k \pm \Delta k$ by momentum conservation, where Δk is the momentum exchange of the scattering process. If the pump laser angle of incidence is chosen at the magic angle, $k - \Delta k$ is very near $k = 0$. In this case, when the pump intensity is increased, the population at $k - \Delta k$ increases superlinearly, because of the Bose stimulated scattering $(1 + n_f)$ factor.

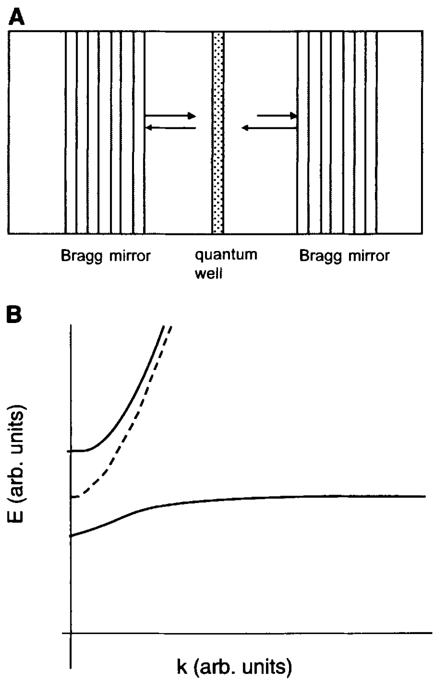

Fig. 5. (**A**) Idealized picture of a semiconductor microcavity. A quantum well is placed between two mirrors, usually composed of multilayer Bragg reflectors. (**B**) A typical polariton dispersion relation in a semiconductor microcavity. In a cavity, the photon dispersion is altered according to Eq. 3, shown as the dashed line in this figure. When the photon energy at $k = 0$ is close to the exciton energy, the mixing of the two states leads to a lower polariton branch, with an energy minimum at $k = 0$.

As a coherent laser pumps the population at k in this case, the populations at $k \pm \Delta k$, which have only undergone one elastic scattering event, retain the coherence of the laser. Therefore, one cannot attribute the stimulated scattering behavior to spontaneous Bose coherence; that is, a thermodynamic phase transition. One can equally well describe these experiments in terms of an optical parametric process (*62*). The two pictures are interchangeable because, as mentioned above, a driven Bose condensate is the same as a coherent classical wave.

Recent experiments (*63*) have shown evidence of spontaneous coherence-starting, using incoherent pumping in which the pump laser is tuned to a k value well above the ground state, so that the polaritons had to undergo several inelastic scattering events before reaching $k = 0$. Earlier experiments of this type (*64*), which were interpreted in terms of spontaneous coherence of polaritons, were later reinterpreted in terms of lasing: stimulated emission of photons instead of polaritons (*65*). The more recent experiments (*63*) have used a distributed set of quantum wells instead of just one quantum well to contain the excitons. In this way, the average polariton density can be kept low even while the photon pumping intensity is high, avoiding the Fermi effects at high exciton density discussed above, which eventually lead to a Fermi level in the excited band and to lasing. The coherence of the polaritons has been measured by photon correlation and shows a gradually increasing degree of coherence as the polariton density is increased.

An intriguing experiment on the spatial propagation of coherent polaritons has been done in a similar system (*66*). Instead of a GaAs heterostructure, two-dimensional layers were made using BiI_3 layers separated by organic barriers. A four-wave mixing experiment was then performed in which two pump lasers were spatially separated, and a third laser probed a spot spatially separated from both. Normally, a fourth wave, which has wave vector and frequency equal to a sum or difference of the other three waves, is obtained only when all three laser spots overlap in the same region of the material. In this case, however, polariton waves from the two pump regions propagated outward and overlapped at the region of the probe laser. The four-wave mixing signal increased superlinearly with pumping density. Again, because a coherent laser created the polaritons directly in a coherent state, one cannot say that the polaritons underwent spontaneous coherence, but these experiments show that the coherent polariton wave will remain coherent over macroscopic distances because of stimulated scattering.

Concluding Remarks

The long-range transport of coherence in exciton or polariton states is one of the tantalizing possibilities of Bose effects in these systems. Although the theory of excitonic and polaritonic condensates is well established, most of the experiments done so far do not provide a direct measurement of spontaneous coherence. As the above survey shows, research on excitonic condensates has taken many directions, and many new results have stimulated the field.

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REVIEW

Cavity Quantum Electrodynamics: Coherence in Context

H. Mabuchi* and A. C. Doherty

Modern cavity quantum electrodynamics (cavity QED) illuminates the most fundamental aspects of coherence and decoherence in quantum mechanics. Experiments on atoms in cavities can be described by elementary models but reveal intriguing subtleties of the interplay of coherent dynamics with external couplings. Recent activity in this area has pioneered powerful new approaches to the study of quantum coherence and has fueled the growth of quantum information science. In years to come, the purview of cavity QED will continue to grow as researchers build on a rich infrastructure to attack some of the most pressing open questions in micro- and mesoscopic physics.

Every real physical system is an open system, and open systems are never more than partially coherent. At its conceptual core, quantum mechanics takes coherence for granted; fully accounting for real-world quantum phenomena thus requires a lot more than straight-ahead integration of the Schrödinger equation. Modern research on open quantum systems scrutinizes this gap between axiomatic theory and empirical realism, and it seeks to clarify murky issues in mesoscopic physics such as decoherence and the emergence of semiclassical dynamics. Cavity QED has long been a central paradigm for the study of open quantum systems and plays a leading

Department of Physics, Mail Code 12-33, California Institute of Technology, Pasadena, CA 91125, USA.

*To whom correspondence should be addressed. E-mail: hmabuchi@caltech.edu