

# Excitons in crystals

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## Abstract

Excitons are elementary electrical excitations of crystals. They are bound states of negatively charged electrons and positively charged holes, which makes them neutral composite bosons. They can exist in either tight- (Frenkel) or weak- (Wannier) binding conditions. The most important property of excitons is to be created by photon absorption and to emit photons when they annihilate. This makes optical methods most appropriate for their studies. They constitute driven-dissipative quantum systems characterized by a rich variety of optical, spin, transport and transfer phenomena. They are important for both fundamental and applied aspects of condensed-matter physics.

## Key points

- The concept of exciton is central to condensed-matter physics.
- Excitons are among the oldest quasi-particles identified and studied, both theoretically and experimentally.
- Excitons are involved in a broad and varied range of effects, mainly optical ones but not exclusively.
- Excitons are expected to undergo a rich set of quantum phases, from Bose-Einstein condensation to superfluidity passing by quantum crystals, Mott insulators and other nontrivial topological phases.
- Excitons remain complex objects even at the two-particle level, whose properties, role, description and sometimes even existence remain hotly debated.
- There is an increasingly thriving activity on excitons thanks to technological progresses and the emergence of new materials.
- There are rich prospects for unique future applications based on excitons, such as stretchable screens.

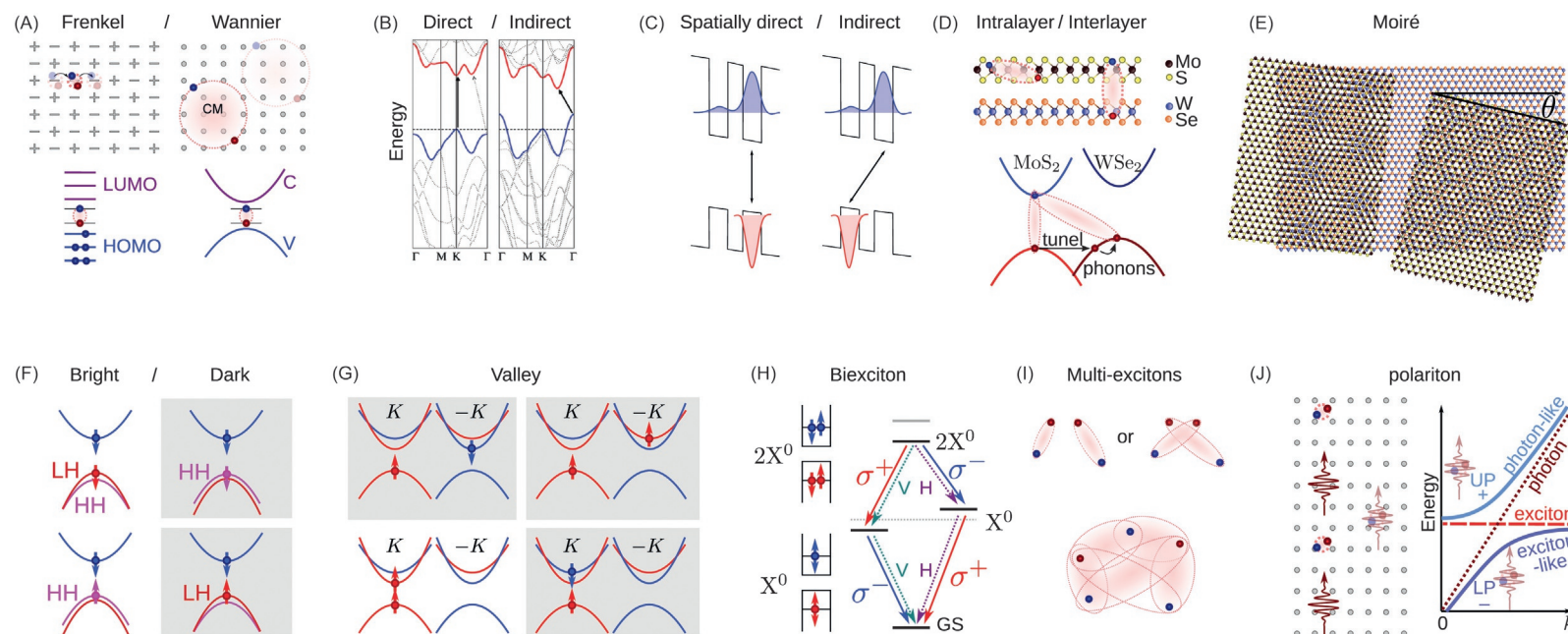
## Introduction

### Historical background: Frankel & Wannier-Mott excitons

The concept—and name—of exciton comes from the soviet theorist [Frenkel \(1931\)](#), starting with his theory for the excitation of atomic states when they are part of a lattice. Excitations in the form of motion or vibrations (sound or heat waves) were already established, by Debye, Tamm, Born and others. Frenkel—who incidentally also named their quantum version as “phonons”—considered instead the case of neutral electronic excitation, that is to say, in raising in energy the quantum state of a single atom in an otherwise frozen (non-vibrating) crystal. The localization of such an excitation does not correspond to a stationary state of the crystal and it is instead diluted over all the atoms in the form of what Frenkel first termed an “excitation wave.” The excited state is then shared by other atoms of the crystal, propagating in this way energy yet with no transfer of charge, as well as momentum yet with no transfer of mass. These running exciton states are the proper stationary states of the static crystal. Such a Frenkel exciton thus describes the situation of an excitation that is not associated with a particular atom, but is travelling from one to another, yet remaining in itself of a size or extent commensurable with the atom. This corresponds to a tight-binding case. Frenkel arrived to the conclusion that the absorption spectrum of a crystal, when not affecting its vibrational state, yields single lines that correspond one-to-one to the absorption lines of an isolated atom of the crystal. This surprising result (at the time) explained why the absorption spectra of solid bodies, which at ordinary temperatures are continuous, break down at low temperatures into spectral lines, as is the case for gases. Such paradoxical observations had indeed been made with the rare earth and iodine spectra, while at higher temperatures, vibrational motion blur this excitonic structure into a continuum. The quantization of such waves and their group packets have been interpreted by Frenkel as giving rise to a particle whose motion is characterized by a wavefunction similar to that of an electron, with a momentum  $\mathbf{p}$  and kinetic energy  $p^2/(2m^*)$  where  $m^*$  is an effective mass. Frenkel named this particle an exciton. A record is found for instance in his 1959 textbook “Introduction to the Theory of Metals” where he writes “*This originating state somewhere does not stay in the place where it was formed but moves uniformly along this or that row of atoms as some particle which I call “an exciton” (“an excitation quantum”).*” Frenkel thus developed a theory of light scattering in crystals that was an analog to Schrödinger’s theory of the Compton effect (scattering of light by a free electron), the electron wave being replaced in his case by his excitation wave. In dielectrics, this also explains why there is no photoconductivity despite the fact that two charges are created in the band model. Pekar reported an interjection during a seminar where this puzzle was being entertained, where Frenkel, after asking why the same phenomenon was not a mystery in gases, simply advised to “*consider a dielectric as a compressed gas.*” This is the foundation of exciton physics. In [Frenkel \(1931\)](#)’s own visionary words: “*the development of the theory sketched above in the quantitative direction [...] does not present any difficulty, for this calculation can be easily reduced to the corresponding calculation for an isolated atom or molecule.*” This fact that characteristics and fingerprints of isolated atoms can survive when the atoms themselves dissolve into an object of an altogether different nature—a solid with a band structure—is an embodiment of the basic idea of *quasi-particles*, the tenet of condensed-matter physics. This similarly allows an electron in the conduction band as well as its absence in the valence band (the “electron-hole,” or “hole” for short) to be treated as negative and positive particles, respectively. This is discussed in more details by [La Rocca \(2022\)](#) in this volume. Beyond the formulation of the exciton as a quasi-particle, Frenkel also analyzed excitons’ interactions with the environment, localization and their optical characteristics. [Frenkel \(1936\)](#) later generalized critically his previous results in order to account for arguments from [Peierls \(1932\)](#) regarding weak excitations, producing what he called “small” excitons, which are now known as *Frenkel excitons*. Since they exist predominantly in molecular (organic) crystals, they are also called “molecular excitons.” Several molecules enter the elementary cell of such crystals, and any of them may be optically excited. The other regime, also anticipated by Frenkel who called them “megaexcitons,” occurs in semiconductors, for instance, in copper peroxide, a popular material of practically all the semiconductor experiments of the 1930s, as well as in the other major classes of semiconductors: germanium, gallium arsenide and the like. [Wannier \(1937\)](#) made a general theory for excitons in the Bloch band continuum, and recognized the case where “*the electron does not escape its hole,*” thus producing no current, despite the fact that the bound electron and hole could be considerably separated in space. They, however, remain correlated, maintaining in particular a well-defined wavevector for their joint propagation. Such large-radii excitons are now known as *Wannier excitons*, and constitute the bulk of excitonic physics in optoelectronics, cf. [Fig. 1A](#). While it is Wannier who derived the Schrödinger equation for the Hydrogen atom for such excitons, it is [Mott \(1938\)](#) who painted the explicit picture for them of an electron and hole bound to each others by the Coulomb interaction in a medium with dielectric constant  $\epsilon$  and migrating around their center-of-mass. This remains, by far, the predominant description of excitons. For this reason, such excitons are also known as *Wannier-Mott excitons*. While his name did not remain attached to excitons, Peierls was also one of the fathers of their conception.

### The existence of excitons

Frenkel’s seminal idea was first received reservedly by the authorities of the time, in particular by Pauli who deemed it to be outright “falsch” (hence not trivial, was Frenkel’s rebuttal). There remained controversy for a while until it became apparent, with [Slater and Shockley \(1936\)](#)’s analysis, that the result was robust to alternative theoretical treatments. The existence of isolated spectral lines, in particular, was drawing attention, since the band-theory of solids precisely dissolves the isolated quantized levels of atoms into continuous bands, so their resurgence was suspicious. Interestingly, modern onlookers of such lines have also raised doubts as to their systematic connection to actual excitons in the system. [Kira et al. \(1998\)](#), for instance, regard this question as a many-body problem of Coulomb-interacting fermionic electrons and holes, which, instead of making transitions between sets of excitons, involve polarization fields. The quantum-mechanical luminescence equations of the electron-hole system happen to produce excitonic resonances, either in absorption or luminescence, even when there is no exciton population in the model. This understanding insists on the difference between the excitonic *polarization* and the excitonic *population*. The former is a complex-valued correlator that probes the transitions permitted by the system, is a coherent quantity subject to dephasing and is



**Fig. 1** Basic concepts of excitons: (A) **Frenkel and Wannier excitons**, attached to one site of the crystal or involving widely separated sites, which translates in the energy diagrams as occupying Highest Occupied (HOMO) and Lowest Unoccupied (LUMO) Molecular Orbitals in one case and living in the band structure in the other. As a result, these two types of excitons are of small and large spatial extension, respectively. Both are, however, delocalized throughout the entire crystal (represented as ghostly images), (B) **Direct and indirect excitons** from negligible or large exchanged momentum in the interband transitions, allowing or preventing an optical process (emission or absorption). Here the character of the transition changes from direct in a MoS<sub>2</sub> monolayer (left) to indirect in its bulk, (C) **Spatially direct and indirect excitons** in coupled QWs. The horizontal axis is the growth direction and the energy levels can be biased by an electric field, with electron (up, blue) and hole (red, bottom) wavefunctions localized in one or the other QW, resulting in single-well excitons or, in contrast, little-overlapping wavefunctions excitons with strong dipole moment and long lifetimes, (D) **Intra and Interlayer excitons** in bilayers of 2D-material TMDs (e.g., MoS<sub>2</sub> and WSe<sub>2</sub>) implement similar ideas than traditional heterostructures in new generations of materials, (E) **Moiré excitons** occur in 2D-heterostructures for various twisting angles  $\theta$  between the two sheets, resulting in potentials of varying lengths and geometries. This new degree of freedom is one of the peculiarities of the new 2D materials, (F) **Bright and Dark excitons** are those which, for various reasons, can and cannot couple to light, respectively. A typical factor is conservation of angular momentum, ruled by the spin of the particles, with only spin  $\pm 1$  excitons recombining (or being created) with a transfer of angular momentum to (from) the photon. This is ruled by the spin of the light-hole (LH) and heavy-hole (HH) bands, (G) **Valley excitons** arise in so-called valleys in the bandgap, which are minima that occur elsewhere than at the zero-momentum (T) point, typically at the  $\pm K$  points on the border of the Brillouin zone. In addition to their spin (red, spin-up; blue, spin-down), electron-hole pairing from different valleys results in momentum-forbidden transitions, leading to more bright/dark configurations (dark shaded in gray). Filling up the bands with more electrons and holes leads to charged valley excitons. (H) **Biexcitons** are the bound states of two electrons and two holes, or two excitons. A biexciton in a QD is sketched with its cascade toward the ground state leading to polarization-entangled photons, which can be hindered by the fine-structure splitting between excitonic states, (I) **Multiexcitons** represent the complicated problem of the identity and description of multiple excitons. Even at the two-exciton level, the indistinguishability of electrons and holes make the formulation in terms of excitons ill-defined. In the presence of even more excitons, charged multi-excitons, trions, quatrions, etc., provide convenient mental pictures which may, however, require independent treatment, (J) **Exciton-polaritons** occur from the strong-coupling between excitons and, e.g., photons, leading to the disappearance of these states as they Rabi oscillate from one type to the other, to give rise to quantum superpositions of a mixed new quasiparticle, in-phase (UP) and out-of-phase (LP) and with new energies which result in a characteristic anticrossing.

not ruled by thermodynamics, while the latter is a real-valued quantity referring to real states (or quasi-particles), subject to relaxation dynamics, equilibrium (if given enough time) and phase transitions. Chatterjee et al. (2004) also find that a plasma of unbound electrons and holes emits at the exciton resonances. Such distinctions are important when it comes to seeking new phases of matter, such as Bose condensation, of particles that may in fact not even be present in the system. The persistence of excitonic lines in a variety of scenarios make the determination of the actual origin a tricky one, with the possibility that the parameter range in which true exciton populations exist is in fact limited despite their ubiquitous manifestation. This conclusion has been amply debated (with as yet no consensual resolution), for instance by Deveaud et al. (2005), although dedicated experimental investigations, such as those by Hägele et al. (1999), tend to confirm that the excitonic problem is indeed more complicated than a bound electron-hole pair popping up and recombining. It could, instead, be explained by much-less intuitive Coulomb-correlation contributions to the photon-assisted recombination of free electron-hole pairs. Kaindl et al. (2003) have sought alternative signatures than spectral lines to evidence the presence of real excitons, including “quasi-particle spectroscopy,” which consists in probing transitions between their Coulombic internal exciton levels, prominently between their 1s and 2p states, which typically falls in the THz range for direct-gap semiconductors. They concluded to a coexistence of excitons with electron-hole pairs but with an ultrafast (quasi-instantaneous) excitonic formation at lower temperatures. Hägele et al. (1999) proposed to measure the electron-hole exchange energy which, when the exciton is bound in the state  $\psi$ , is proportional to its electron and hole overlap  $|\psi(0)|^2$ . This can be obtained from the quantum beating of spin since the exchange energy, when it is present, turns the linear dependence of its frequency on an applied magnetic field into a parabolic one (due to a correction to the Zeeman splitting). They concluded that a dominant population of excitons is present only under stringent experimental conditions. Progress from material science, providing more robust excitons, is helping to bring back an unanimous recognition of excitons. In TMDC materials for instance, excitons are believed by Steinhoff et al. (2017) to prevail over electron-hole plasma even up to room temperature, through balancing processes of a fission-fusion type between the two types.

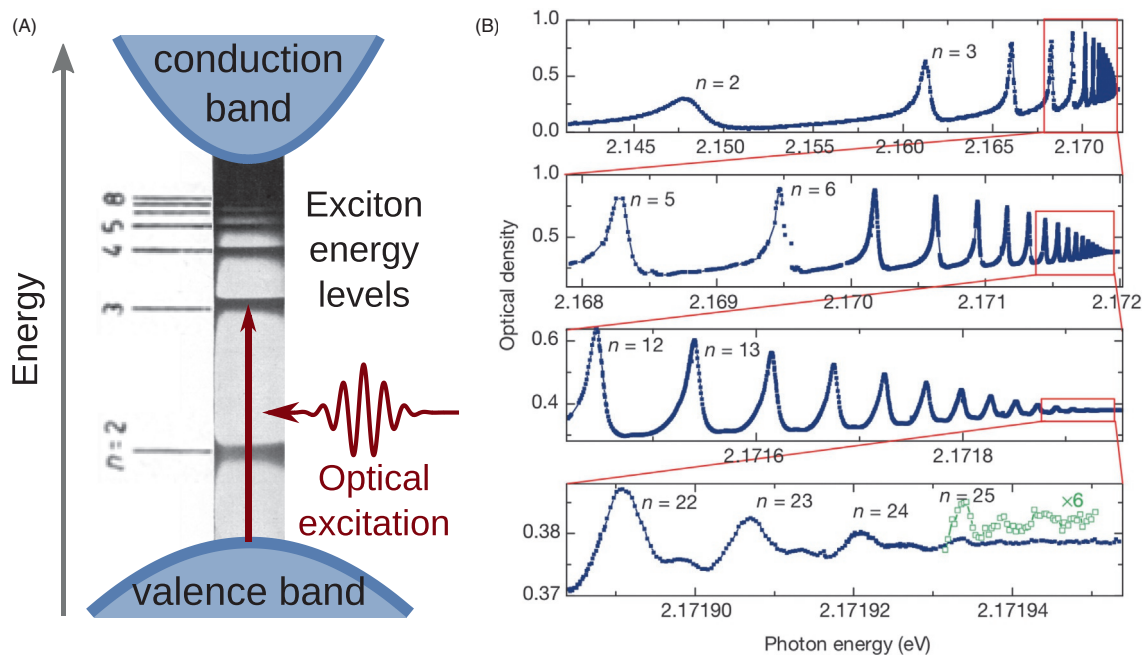
Another hotly debated problem with excitons is whether they qualify, or even exist, as bosons. The commutator of the annihilation and creation operators of excitons is not 1 but deviates from this bosonic limit by an order  $na_B^D$  in  $D$  dimensions, giving hope of behaving as good bosons at low enough densities. The composite nature of excitons, however, combined to their large size and delocalized character has been hailed as voiding their validity as interacting bosons already at the two-particle level. Since electrons and holes cannot be attributed to any single exciton, one should deal directly with a fermionic four-particle complex dominated by the Pauli exclusion principle. Illustratively, the interaction of any electron with any hole is part of the exciton-exciton scattering when these particles are assumed to belong to different excitons, but are also constitutive of the excitons themselves when belonging to the same exciton, in which case they should be ignored since they have already been taken into account to bind the exciton in the first place (Fig. 11). Combescot and Betbeder-Matibet (2002), in particular, argue that purely fermionic contributions are of the same order as Coulomb contributions and, more importantly, forbid an effective Hamiltonian, since two-exciton interactions involve all other excitons. This is because Pauli exclusion is intrinsically a  $N$ -body effect, which precludes an exact description with a standard two-body interactions, even with some inclusions of the exchange terms. They observed, for instance, that such an ad hoc procedure produces a non-Hermitian Hamiltonian. They conclude that the conventional “bosonization”—which consists in mapping the original fermion problem into one of elementary bosons so as to apply known and familiar techniques—are erroneous and should not be used, but replaced by a many-body theory of “cobosons” (for composite bosons), such as that exposed by Combescot et al. (2008). The extent to which the composite nature of excitons affects the dynamics of several excitons remains a problem of active research.

## Overview of excitonic physics

### Theory of excitons

Excitons are primarily a theoretical concept: a useful physical picture to capture the essence of a phenomenon which, in its full complexity, fails to adhere exactly to any but a complete, so-called “ab-initio” or “first-principle,” brute force calculation of the Schrödinger equation for the full system with no phenomenological parameters. Although ab initio methods are better suited for ground states calculations (with density functional theory), they can also compute elementary excitations in a self-consistent way and derive optical response. A so-called GW ( $G$  is for “Green function” and  $W$  refers to screening of the Coulomb interaction) approximation combined to the Bethe-Salpeter equation (describing bound states in quantum field theory, so particularly suitable for excitons), or GW-BSE approach, has proved particularly accurate to reproduce experiments in great details. In such cases, one needs not speak of excitons but focus on the calculated observables and compare them to experiments. The agreement in some cases can be extremely good. Even such approaches remain however limited by computational resources and need to make some approximations, that have to be both understood and controlled. Various models thus derive various excitons in different limits. Frenkel and Wannier excitons are obtained in this way from the same underlying theory but using different basis functions. They both start with the Born-Oppenheimer approximation, that decouples the electrons and ions dynamics so that Schrödinger’s equation can now focus on the electronic wavefunction  $\phi(\mathbf{r}_1, \mathbf{r}_2, \dots)$  alone, i.e., for the joint probability amplitude to find the  $N \gg 1$  electrons in the crystal at positions  $\mathbf{r}_i$ . If the crystal is assumed static (frozen atoms), the Hamiltonian retains electrons’ kinetic energies  $-\frac{\hbar^2}{2m} \sum_i \nabla_i^2$  ( $m$  the electron mass and  $\nabla_i$  acting on the  $i$ th variable of  $\phi$ ), the electron-crystal Coulomb interaction  $-\frac{1}{4\pi\epsilon} \sum_i \sum_n Z e^2 / |\mathbf{X}_n - \mathbf{r}_i|$  and electron-electron interactions  $\frac{1}{2} \frac{1}{4\pi\epsilon} \sum_i \sum_{j \neq i} e^2 / |\mathbf{r}_i - \mathbf{r}_j|$ , with  $Z$  the number of charges of each atomic site,  $e$  the electron charge,  $\mathbf{X}_n$  the positions of the atoms, that define the crystal and  $\epsilon$  the dielectric constant. Since each electron interacts with every atom of the crystal and with every other electron, the problem, although simply formulated, is extremely complicated. The next popular step is the one-electron approximation that breaks  $\phi$  into a product  $\phi(\mathbf{r}_1, \mathbf{r}_2, \dots) = A\varphi_1(\mathbf{r}_1)\varphi_2(\mathbf{r}_2)\dots$  with  $A$  the antisymmetry operator that changes signs whenever two electrons are permuted, to enforce their Fermi-statistics. This

factorization makes the electrons uncorrelated, which is clearly an oversimplification, but correlations are recovered by linear superpositions of such states that serve as basis functions, and which have been introduced in this form to decide on an explicit form. Two popular choices—tight-binding and Bloch functions—lead to the two main types of excitons: Frenkel and Wannier. With tight-binding functions, the electron remains localized to its atom, as is the case in organic solids where there is weak overlap between lattice sites due to small intersite interactions. Bloch functions, on the other hand, have the periodicity of the crystal built-in as  $\varphi_j(\mathbf{r}_j) \equiv u_{\mathbf{k}}(\mathbf{r}_j) \exp(i\mathbf{k}_j \cdot \mathbf{r}_j)$ , introducing a momentum  $\mathbf{k}_j$  as a label of states and with  $u_{\mathbf{k}}(\mathbf{r}_j + \mathbf{R}) = u_{\mathbf{k}}(\mathbf{r}_j)$  for any vector  $\mathbf{R}$  connecting two sites of the crystal. They thus delocalize the electron in the solid thanks to the band structure. In both pictures—bound and propagating electrons—the exciton that arises from exciting one electron from any one site, is propagating (i.e., is delocalized), like the Bloch electron. Frenkel and Wannier excitons are thus two limiting models of the same theory, cf. Fig. 1A. The Wannier model is typically valid for low-lying exciton states in semiconductors and crystals with a large dielectric constant. It breaks down for small-radius excitons because first-order corrections are large and hard to calculate, and also because small distances mean a large spread in momentum and thus sensibility to the non-parabolic band structures. A beautiful feature of the Wannier treatment is the natural emergence of Schrödinger's equation for the hydrogen atom for the electron-hole pair. From this, follows straightforwardly the energy of an exciton in a semiconductor crystal as  $E_X(\mathbf{k}) = E_{\text{gap}} - R_y + \frac{\hbar^2 \mathbf{k}^2}{2m} + \dots$  where  $E_{\text{gap}}$  is the bandgap energy, and  $R_y$  is the binding energy. The latter is known from the hydrogen atom as the Rydberg energy  $R_y \equiv \frac{e^4 \mu_0}{2\epsilon^2 \hbar^2}$  with  $\mu_0 \equiv m_e m_h / (m_e + m_h)$  the reduced mass computed from the electron  $m_e$  and hole  $m_h$  effective masses. These masses correspond to a quadratic fit to the dispersion relation of the exciton as given by band theory, and can be negatives. This connection to atomic physics is a central manifestation of excitons in crystals, as shown in Fig. 2. The excitonic Bohr radius is also obtained from the atomic theory as  $a_B = \frac{\epsilon^2 \hbar^2}{\mu_0 e^2}$ . Therefore, one can relate the size of the exciton, as estimated by its Bohr radius, to its binding energy as  $R_y = \frac{e^2}{2\epsilon a_B}$ , i.e., chiefly through the dielectric constant  $\epsilon$ , that can vary a lot from one dielectric to the other. The dielectric constant, also called “relative permittivity,” quantifies the polarizability of a dielectric and how this affects the Coulomb force between two point charges in the medium as compared to the vacuum. This is frequency dependent, due to retardation effects (the polarization of a material does not follow instantaneously variations of an electric field). This results in phase shifts that make the dielectric constant complex-valued. The short-range interaction is governed by the low frequency limit of the dielectric constant, while the long-range interaction is ruled by its high (optical) frequencies. Since the Coulomb potential is central to the electron-hole interaction, the features of the dielectric constant impact directly the structure of the exciton. For instance, the dielectric constant is typically large in semiconductors, resulting in important screening of the interaction and thus a weak binding energy, or, in turn, large excitons. Frenkel and Wannier excitons therefore typically occur in small and large dielectric constant materials, respectively, although this admits exceptions. Dimensionality is another crucial factor for the dielectric constant and thus also for the exciton. First Rytova (1967) and Stern (1967), independently, (later, also Keldysh (1979) and others) derived the screened Coulomb potential in two-dimensional structures, featuring a logarithmic shape at short distances and a hyperbolic shape at long distances.



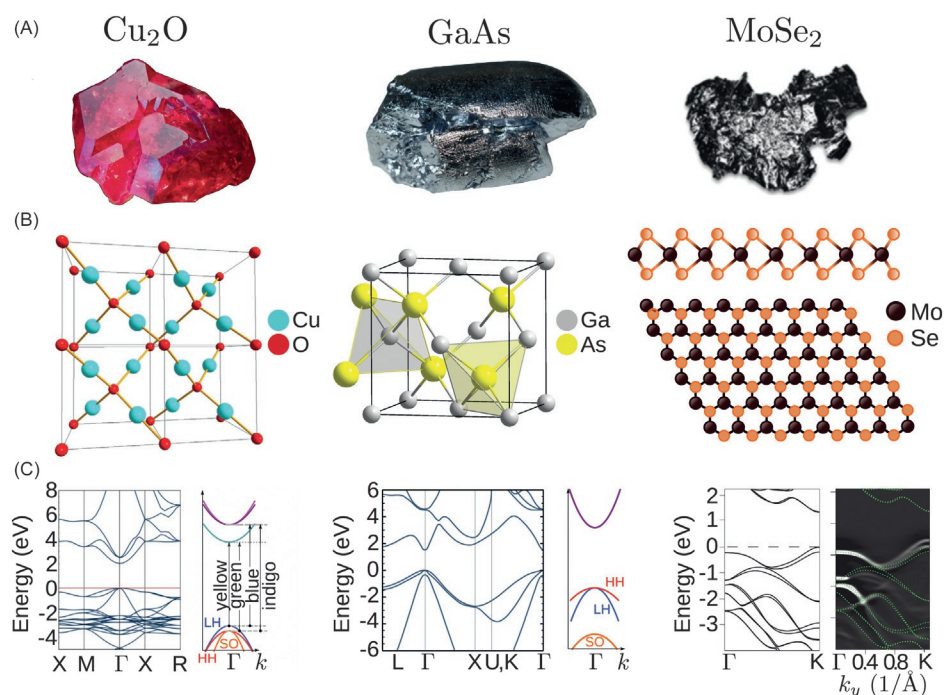
**Fig. 2** Optical excitation of an exciton in the semiconductor cuprous oxide: the spectral lines are observed in the bandgap due to the negative (binding) energy of the excitonic states. In (A), as first observed by Gross (1956) (up to  $n = 8$ ) and in (B), as best observed by the Manfred Bayer group (Kazimierzczuk et al., 2014) with very-high (Rydberg) exciton states (up to  $n = 25$ ). The same material, Cu<sub>2</sub>O, shows the impressive progress of technology in the course of a half-century. (A) From Gross EF (1956) Optical spectrum of excitons in the crystal lattice. *Il Nuovo Cimento* 3:672, <https://doi.org/10.1007/BF02746069>. Panel (B) is reproduced with permission from Kazimierzczuk T, Fröhlich D, Scheel S, Stolz H, and Bayer M (2014) Giant Rydberg excitons in the copper oxide Cu<sub>2</sub>O. *Nature* 514:343.

In one-dimension, one can encounter large-radius excitons with large binding energies, i.e., of a mixed Frankel–Wannier character, due to the shape of the Coulomb potential in such a case (large dielectric constant but small screening). Another potential of note was introduced by Haken (1956), to describe the electron-hole interaction screening in an oscillating lattice, which occurs with phonon vibrations. This results in a distance-dependent dielectric function. In all cases, the binding energy also quantifies the exciton's stability with temperature, up to  $T \approx R_y/k_B$  (through Boltzmann constant). The hydrogenic theory also provides a suitable framework to study excitonic responses to applied external fields, in particular through perturbation methods. Understood as huge atoms, one can indeed account for a wealth of properties of excitons that are well-known from atomic physics, such as their easy ionization (with relatively weak electric field as compared to hydrogen), or, before this happens, the Stark effect (shift of the spectral lines under an applied electric field) that is observed with hundreds of volts only, or the diamagnetic effect, Zeeman splitting, Landau levels, etc. Most of these observations have been made in the early years of the field, mainly by Gross (1962) and coworkers (in particular, Zakharchenya) in the wake of their observation of the spectral structure. Littlewood (2022) provides a more detailed exposition of the theory of excitons in this volume.

### Excitons in various materials

Excitons appear in solids of various types: in molecular, ionic (particularly the alkali halides), and semiconductor crystals, and in complicated organic compounds. Excitons are too-short lived in most metals to be observable but they occur in superconductors. They have also been observed in some liquids, and even in living matter, where their migration is believed to be an important mechanism of energy transport for photochemical reactions such as photosynthesis. At a more basic level, exciton generation, diffusion, and annihilation are known to be involved in the efficiency of photovoltaic solar cells, although processes there are typically too complex to be described in these terms. It is definitely in crystals that excitons are currently the best understood and the most actively studied. Common properties are found in all cases which can be seen as those defining the exciton (such as the hydrogenoid spectrum  $-R_y/n^2$  for Wannier excitons). Still, the details of each solid make their corresponding excitons be sui generis objects. We now give a broad overview of how various materials give rise to various excitons (cf. Fig. 3).

One crystal of choice for excitons is the one in which their first compelling observation was made, namely, cuprous oxide,  $\text{Cu}_2\text{O}$ . This is one of the main oxide of copper (the other being cupric oxide  $\text{CuO}$ ) through the process  $4\text{Cu} + \text{O}_2 \rightarrow 2\text{Cu}_2\text{O}$ . This red (sometimes yellowish) semiconductor remains to this day—surprisingly, in its natural form even better than when



**Fig. 3** Emblematic materials hosting excitons:  $\text{Cu}_2\text{O}$ , GaAs and  $\text{MoSe}_2$ , with comparisons of (A) the bulk crystals (an exfoliated foil for  $\text{MoSe}_2$ ), (B) the crystal structures (a mono-layer as seen from the side and from the top for  $\text{MoSe}_2$ ) and (C) the band structures. For  $\text{Cu}_2\text{O}$  (after Meyer et al. (2013)) and GaAs (after Richard et al. (2004)), the full calculated bandgap is shown along with the parabolic approximation around the  $\Gamma$  point. For a monolayer of  $\text{MoSe}_2$ , the experimental band structure is superimposed by Zhang et al. (2014a) to the theoretical prediction, showing a stronger direct bandgap than is calculated. Panels (C) are adapted with permissions from Meyer BK, Polity A, Reppin D, Becker M, Hering P, Kramm B, Klar R, Sander T, Reindl C, Heiliger C, Heinemann M, Müller C, and Ronning C (2013) The Physics of Copper Oxide ( $\text{Cu}_2\text{O}$ ). Academic Press, chapter 6. <https://doi.org/10.1016/B978-0-12-396489-2.00006-0> and Zhang Y, Chang T-R, Zhou B, Cui Y-T, Yan H, Liu Z, Schmitt F, Lee J, Moore R, Chen Y, Lin H, Jeng H-T, Mo S-K, Hussain Z, Bansil A, and Shen Z-X (2014) Direct observation of the transition from indirect to direct bandgap in atomically thin epitaxial  $\text{MoSe}_2$ . *Nature Nanotechnology* 9:111.

synthesized—the best one to host the cleanest and finest excitonic states, as illustrated in Fig. 2. The highest valence and lowest conduction bands stem from the copper atoms in the 3d and 4s orbitals, respectively. The exciton formed by the corresponding electron and hole provides the so-called “yellow series” with energies around 2.1 eV. The 1s state has a binding energy of order 150 meV. This lowest exciton state in  $\text{Cu}_2\text{O}$  is optically forbidden but becomes activated under a strong magnetic field and has been resolved in this way by Brandt et al. (2007) as an essentially perfect Lorentzian with a linewidth as small as 80 meV, making it the narrowest bulk exciton resonance ever observed.

Excitons have also been extensively studied in III–V semiconductors. Gallium Arsenide (GaAs) is the prototypical conventional semiconductor for exciton physics. Semiconductors with a large dielectric constant have only weakly bound excitons, which can be observed only at cryogenic temperatures. In GaAs, the binding energy of excitons is 4 meV, so they ionize much before room temperature. In contrast, in GaN, the exciton binding energy is as large as 28 meV, which makes this wide bandgap semiconductor material suitable for exciton studies at room temperature. In silicon and germanium, the mass is not as isotropic as in other semiconductors and the band structure is more complex, requiring heavy calculations to derive the experimentally observed binding energies. II–VI materials, such as ZnO, ZnTe and CdSe, feature much higher excitons with binding energy in the range from 50 to 200 meV, so they are stable at room temperature, but such structures come with their own limitations, such as a higher disorder. II–VI semiconductor alloys containing magnetic ions such as  $\text{Mn}^{2+}$  offer a convenient playground for studies of magneto-excitons and exciton magnetic polarons. Such materials are typically characterized by giant Zeeman splitting of exciton states that would be described by effective exciton  $g$ -factors of the order of 50.

Graphene—a single layer of carbon atoms arranged in a two-dimensional honeycomb lattice—pioneered a new era of 2D crystals. Graphene’s dispersion relation becomes linear and its electrons thus behave as Dirac (massless) particles, with conduction and valence bands that touch on the edges of the Brillouin zone, thus making graphene a semimetal (or gapless semiconductor). For this reason, its excitonic features, while existing, are not compelling. Rolled-up version of graphene—carbon nanotubes—can be either metallic or semiconducting depending on how they are rolled. Even when completely metallic, and thus expected to be even less excitonic than graphene, the strong 1D screening can reduce interactions sufficiently so that metallic excitons have been observed in nanotubes by Wang et al. (2007). Semiconductor nanotubes, on the other hand, sustain a rich variety of excitons, characterized by large oscillator strengths. In lower dimension, another allotrope (structural-variation) of carbon—the so-called carbyne—is strongly believed to exist in the form of a linear carbon chain of the type  $(-\text{C} \equiv \text{C})_\infty$ . Partial chains of this type stabilized at their ends, known as polyynes, are well established and have been shown by Kutrovska et al. (2020) to exhibit sharp excitonic features at low temperatures. In the wake of graphene, material physics developed other similar two-dimensional crystals that are semiconductors, such as hexagonal boron nitride (hBN, known as “white graphene”), molybdenum disulfide ( $\text{MoS}_2$ ), 2D tungsten disulfide ( $\text{WS}_2$ ), tungsten diselenide ( $\text{WSe}_2$ ), molybdenum diselenide ( $\text{MoSe}_2$ ) and many other dichalcogenides and layered oxides. Their excitonic properties are particularly important since 2D crystals are more stable when they are insulators or semiconductors. Transition metal dichalcogenides (TMDs or TMDCs) are those with the chemical formula  $\text{MX}_2$  with M an atom from the transition metal family, in particular Mo (Molybdenum) or W (Tungsten), while X is a chalcogen, i.e., an atom from the group 16 of the periodic table, which is that of oxygen, but, for TMD, with X = S (Sulfur), SE (Selenium) or Te (Tellurium). While bulk versions of these materials are indirect bandgap semiconductors, atomically thin layers acquire a direct bandgap in the near-infrared and visible regions, so of great interest for opto-electronics. This is shown from calculations reported by Splendiani et al. (2010) in Fig. 1B and was established with spectroscopy by Mak et al. (2010). These materials also have exceptionally strong electron-hole Coulomb interactions, with record-value binding energy on the order of several hundreds of meV, thanks again to the reduced screening of the Coulomb interaction in 2D, that also manifests in its excited or charged states. This makes their exciton stable at room temperature. While they are of the Wannier type, their huge binding energy is more characteristic of Frenkel excitons. They also have energy-degenerate band edges at the  $\pm K$  points, at the corners of the Brillouin zone, resulting in several extrema of the band structure, known as “valleys,” whose occupation result in “valley excitons” (cf. Fig. 1G), which differ substantially from excitons in conventional semiconductors whose bandgap is at the center of the Brillouin zone ( $\Gamma$  point). To start with, they come with a large variety of bright and dark configurations, especially when also combining valleys with the electron and hole spins. Optical transitions come with new selection rules, with each circular polarization coupling to one valley ( $+K$  to  $\sigma^+$  and  $-K$  to  $\sigma^-$ ) allowing to easily address each valley by polarization. A strong spin-orbit interaction produces spin splitting of several hundred meV in the valence band and of a few meV only in the conduction band. Valley excitons also behave differently under applied external fields and present great technological prospects. The lowest energy bright valley exciton (known as the “A exciton”), is accompanied by an higher-energy, shorter-lifetime version (the “B exciton”), with the hole residing in the higher energy spin-split valence band, and that decays non-radiatively to the A exciton. There is also a rich collection of charged excitons from the various possible combinations in the valley and split-bands. These valley excitons additionally feature a strong fine-structure due to the exchange interactions between the valleys. While their characterization is complicated, there are also more tools to do so, e.g., polarization-resolved pump-probe and spectral hole-burning measurements.

### Spin structure

The exciton spin state is given by the spin of its constituents: electrons and holes. Being fermions, electrons and holes have semi-integer spins that add up to integer numbers, so that excitons should be bosons according to the spin-statistics theorem. The spin projections of electrons and holes onto the crystal axis are not necessarily  $+\frac{1}{2}$  and  $-\frac{1}{2}$ . As an example, in Zincblende

(two interpenetrating face-centered-cubic lattices) semiconductors such as GaAs or CdTe, the valence band is two-fold degenerate with branches characterized by spin (strictly speaking, an effective spin comprising also the angular momentum) projections of  $\pm\frac{3}{2}$ , referred to as heavy holes, and  $\pm\frac{1}{2}$ , referred to as light holes. In quantum wells, the degeneracy of the valence band is lifted, and the lowest-energy exciton state is formed by a heavy hole and an electron. Heavy-hole excitons are characterized by four possible spin projections on the structure axis:  $-2$ ,  $-1$ ,  $+1$  and  $+2$ . Momentum selection rules only enable exciton states with  $\pm 1$  spin projections to emit or absorb photons, which are known as *bright excitons*, cf. Fig. 1F. In contrast,  $\pm 2$  excitons cannot be directly excited by light and they cannot recombine by emitting a single photon. These optically inactive excitons are thus referred to as *dark excitons*. They play an important albeit implicit role, by interacting with optically active or bright excitons. Spin-orbit interactions for excitons are prominent for the understanding of their optical properties. They stem from two origins: (1) spin orbit interactions for the constituent electrons and holes, such as Eliot-Yaffet, Dyakonov-Perel' and Bir-Aronov-Pikus mechanisms and (2) short- and long-range exchange interactions, which result in the specific fine structure of exciton energy spectra. The longitudinal-transverse (LT) splitting—which is the splitting between exciton resonances in the absorption spectra of light polarized parallel and perpendicular to the in-plane component of the exciton wavevector, in-plane of the quantum well—is caused by the long-range electron-hole exchange interaction. LT-splitting is at the origin of the Maialle-Sham mechanism of spin relaxation for excitons (published by Maialle et al. (1993)), that is similar to the Dyakonov-Perel' spin relaxation mechanism for electrons.

### Optical properties of solids

The most important properties of the most common type of excitons arise from their link to light absorption (creating excitons) and emission (when excitons recombine), thus connecting electrical excitations inside the crystal to photons outside, cf. Fig. 3A. This admits exceptions as excitons can also be formed by electrical injection of electrons and holes, e.g., as implemented by Schneider et al. (2013) to realize an electrically pumped exciton-polariton laser. Nevertheless, excitons play a predominant role in the optical properties of solids. Optical methods are also the most appropriate to study them. The most common form of characterization of excitons is found in the absorption and reflection spectra of insulating crystals. The case of semiconductors is particularly important, due to their rich physics and technological importance. Another successful method for characterizing the excited states of excitons is to create them with photons of energy greater than that of the energy gap and to measure the far-infrared absorption due to direct transitions between excitonic levels. A third experimental technique is to detect the luminescence emitted when the photo-produced exciton recombines. The system can also be probed by two-photon excitation, in which case, not the ground state  $1s$  but the excited  $2p$  couples to light. This allows, among other things, to measure the excitonic energy spectrum and thus to get an estimate of the binding energy. In regions of no absorption, the semi-classical theory of radiation conveniently describes the optical response due to excitons through the dielectric constant of the crystal. In regions of absorption, the quasi-particle picture is favored. In semiconductors with a small binding energy, excitons are the main mechanism of light emission and absorption, although at low temperature only, since at higher temperature, they are not bound anymore and form a plasma of electrons and holes instead. At low-enough temperature, the hydrogen-like spectrum of absorption is observed just below the band gap, which is itself easily identified as an edge in the absorption spectrum. Optical properties are strongly dependent on dimensionality. In bulk, the exciton absorption lines are asymmetric with a characteristic Fano lineshape, described by Toyozawa (1958), which is typical whenever a discrete state (here the exciton) interacts or interferes with a continuum (here optical phonons). The total energy of the exciton is determined by the band-gap energy, the binding energy and the kinetic energy due to its center-of-mass motion. Photoluminescence (PL) can provide information on all these aspects, allowing in particular to extract the distribution of excitons velocity. In 2D, in the absence of excitonic effects, direct band to-band transitions exhibit a step function spectrum that echoes the energy-independent density of states, but in the presence of excitons, the sharp hydrogenoid resonances are observed on top of this structure. In many instances, excitons can also be controlled optically through the quantum coherent phenomenon of Rabi oscillations, i.e., the hermitian cycling that alternates stimulated emission by the exciton of a photon and the reverse absorption of the photon by the exciton. Controlled pulses thus allow to prepare the exciton in a deterministic quantum state, such as a full exciton or a quantum superposition with its ground state. In fact, such coherent light-matter processes are inherent to excitons and give rise to a whole chapter of excitonic physics: polaritonics.

### Exciton-polaritons

Just like a self-consistent theory of the electron demands the electromagnetic field (for gauge invariance) and thus its interaction with light, a theory of the exciton cannot dispense from the electromagnetic field. Hopfield (1958) pointed out that the semiclassical description of light absorption by excitons is not “completely satisfactory,” in contrast to absorption due to interband transitions for instance, because of strong photon-exciton coupling phenomena. These were referred to as “light-excitons” in the soviet literature but Hopfield christened them “polaritons” instead, as a general phenomenon of coupling light with a polarization field, that is not limited to excitons (e.g., optical phonons also provide a polarization field for light). This polaritonic effect implies that in bulk semiconductors, excitons actually impede luminescence and radiation, by trapping light into cyclic (Rabi) oscillations with the excitons (Fig. 1J), which breaks only at the surface so that in an infinite crystal, excitons are non-radiative. Similarly, light is not absorbed by excitons but converted into exciton-polaritons, so that true absorption is the result of other mechanisms, such as exciton-phonon scattering, in the absence of which light would be reflected or transmitted. Quantum mechanics also makes the

exciton-photon coupling relevant even in absence of photons. In particular, the polariton vacuum is not the crystal vacuum. At the heart of such polaritonic effects lies the one-to-one coupling of the modes (electromagnetic and excitonic), so that in a system of reduced dimensionality, luminescence is indeed dominated by free excitons, as shown by Weisbuch et al. (1981). This results from a Wigner-Weisskopf dissipation of one exciton to a continuum of photon modes. To restore strong coupling, one must embed the lower-dimensional crystal in a microcavity that also reduces the dimensionality for photons. This gives rise to cavity-polaritons, that are half-light/half-matter quasi-particles combining the electronic properties of the exciton (strong interactions, large mass, ...) and the ultrafast ones of the photon (high coherence, small mass, ...). This new brand of quasi-particles has been revealed in optical experiments by Weisbuch et al. (1992) in two dimensions, by Tartakovskii et al. (1998) in one dimension and by Yoshie et al. (2004) and Reithmaier et al. (2004) in zero dimension. Unlike bulk polaritons—that are tricky to monitor, let alone controlled, due to their dynamics being buried inside the material until they reach its borders—cavity polaritons can be conveniently and continuously monitored thanks to the leakage of the photon from the cavity. A typical degree of freedom in such systems is the exciton/photon fraction of the polaritons, quantified by the Hopfield coefficients that act as a probability amplitude for the quantum superposition between the photon and the exciton. This can be tuned by matching the dispersions of the two modes, so that the excitation can be smoothly changed from one extreme (exciton) to the other (photon) passing by equal superpositions. Even at resonance, where the admixture is half-light, half-matter, different excitation schemes can yield exciton-like or photon-like polariton dynamics in dissipative systems. This results in qualitative differences of their luminescence from a coupled-oscillator model, that otherwise simply links the splitting of the lines to the Rabi coupling, as discussed by Laussy et al. (2008). Thanks to their even more strongly driven-dissipative character, polaritons have rapidly grown from an annex of exciton physics to rapidly expanding and independent research fields, which are overviewed by Kavokin et al. (2017) and discussed in more details in this volume by Litinskaya (2022).

### Excitons interactions

Excitons interactions concern the problem of an exciton in presence of other particles or quasi-particles, either other excitons, in which case one deals with self-interactions, or with other types, such as photons (as discussed previously), electrons, holes, phonons, plasmons, etc. Excitons can also interact with (scatter off) defects. Each interaction constitutes a field of research on its own. The basic picture of quasi-particles allows one to embrace the main ideas at a glance.

The exciton-phonon interaction is both important and enlightening as a particular case. It involves the exciton in the more basic problem of electron-phonon coupling, which can itself be described by a variety of models. Prominent electron-phonon Hamiltonians include the Fröhlich and the Holstein Hamiltonians. Exciton-phonon scattering amplitudes can be computed from them with Fermi's golden rule applied to the corresponding exciton-phonon matrix elements. Physically, the presence of an exciton modifies the charge configuration within the crystal by shifting the equilibrium positions of the lattice ions, which gives rise to a coupling between excitons and lattice phonons. Depending on the type of phonons, the interaction can be short range (acoustic phonons) or long range (optical phonons). When such interactions are very strong, and when the hole and electron are greatly separated, they may polarize the lattice individually, the result being a polaron bound to a hole-polaron (the polaron itself being the particle that arises from strong electron-phonon coupling). The "*exciton-polaron*," on the other hand, refers to strong electron-exciton coupling. Strong phonon-exciton coupling can also cause self-trapping, whereby small-radius excitons are clouded by phonons which impede their ability to move. Phonons are affecting a broad range, if not all of the excitonic behaviors, including their energy, oscillator strength, lifetime, coherence, relaxation dynamics, diffusion, luminescence and spectral shapes. In all cases, this can be identified by a temperature dependence. Phonons also open a new (so-called non-radiative) decay channel, which increases with temperature and also causes broadening of the spectral lines. The other way around, phonons are also required to bind an exciton from a pre-existing free hole and free electron, to conserve energy and momentum. They also couple bright and dark excitons. Ab initio Bethe-Salpeter calculations by Marini (2008) in two-limiting cases of weak (in bulk silicon) and strong (in hexagonal Boron nitride) exciton-phonon coupling reproduced accurately the thermal properties of these systems, and found that phonons cause substantial changes even at zero temperatures, such as renormalization (weakening) of their binding energy. Joint incoherent contributions—with independent interactions from the electron and hole of the exciton with the lattice vibration, dominant at weak coupling—and coherent ones—affecting the exciton as a whole and in particular contributing to its buildup at stronger coupling—were identified. Also, a microscopic mechanism for bright and dark excitons transitions was found, based on a transfer of optical strength between energetically-close excitons. Overall, such successful simulations of the excitons-phonons interactions show both their variety and importance in realistic systems, even at low temperatures. Other approaches, for instance based on a master equation formalism, have similarly captured counter-intuitive results and qualitative departures of excitons from atomic systems due to their solid-state environment and coupling to phonons, such as McCutcheon and Nazir (2013)'s finding of an increased quantum coherence due to thermalization of the dressed states.

If there is one basic problem for which the hydrogen body of knowledge brought little insights when applied to its excitonic version, this is the fundamental one of exciton self-interactions. The similarity of the masses of all particles is a first departure from the hydrogen atom. The delocalization of the exciton throughout the crystal, i.e., involving all electrons and holes, is another crucial difference that makes two excitons intrinsically overlapping even when neglecting their interactions, which is the existential problem of cobosons previously discussed. For 1s excitons, since they are electrically neutral, their Coulomb interaction is small, even though largely separated excitons can experience a van der Waals interaction. Exciton interactions become notable when the two excitons get in close proximity to each other. In this case, exchange interactions of the constituent electron and hole become

dominant while the direct Coulomb interaction remains small. The exchange interaction is due to the antisymmetry of the fermionic wavefunction, that tends to push particles apart, as opposed to bosonic symmetry that produces an attraction. The simplest, because the most familiar, and for this reason also the most popular treatment of exciton-exciton interactions is through their bosonization, in which case they are assumed to be structureless (ideal) bosons. Although this procedure, introduced by Hanamura (1974) and others, is still of unclear accuracy or even validity, it appears to provide satisfying result to account for a wealth of experimental results. These include a blueshift of the exciton energy with increasing excitation, a saturation of the oscillator strength as well as collisional broadening, or homogeneous increase of the linewidth. Ciuti et al. (1998) computed scattering strengths (in 2D as befits quantum wells) taking into account the exchange terms, which are the dominant ones. Besides, they differ qualitatively from the direct Coulomb term, being maximum ( $\approx 6E_B a_B^2/S$  with  $S$  the surface area) for no-exchanged momentum while the Coulomb interaction vanishes in this case. Spin also plays an important role in exciton interactions, with possible scatterings into dark states. Depending on the relative spin, both attraction and repulsion between excitons are possible. Excitons with different spins are distinguishable and thus the dominant exchange terms do not apply to them. In this case, another strong channel of scattering involves the bound state known as biexciton, which we discuss next. While the problem of exciton interactions in its full generality remains a complex area of hotly disputed research, basic consequences such as bound states have gained wide recognition.

### Multiexcitons

When several excitons bind together, they form a multiexciton (cf. Fig. 11). The most popular multiexciton is the biexciton, the quasiparticle composed of two electrons and two holes and that constitutes the solid counterpart of a molecule (Fig. 1H). This complex may be stable thanks to the dipole-dipole attraction of two excitons polarized in opposite directions. The spin structure of the constituent electrons and holes is thus essential in this formation. Usually, only singlet states formed by excitons with opposite spins are stable. This imposes a polarization selection rule: biexcitons cannot be directly optically excited by a circularly polarized light as it would only generate excitons with parallel spins that repel each other because of the exchange interaction. In contrast, biexcitons can be excited by a linearly polarized light capable of generating excitons with opposite spins. As the excitation of each biexciton requires two photons, the probability of their generation increases quadratically with the intensity of pumping light, which is a typical signature of their formation. The two-photon excitation energy should be lower than that of two unbound excitons due to their binding energy and spin interactions (higher if these turn out, for structural reasons, to exceed two-excitons energy). Other complexes exist. Trions consist of positively or negatively charged excitons composed by two holes and one electron or two electrons and one hole, respectively. They can be excited in doped semiconductor structures containing residual electrons or holes: an optically excited electron-hole pair binds itself to a residual carrier giving rise to the trion formation. Trion binding energies in bulk semiconductors are usually very small because of the little effective mass imbalance between electrons and holes. In quantum confined structures, such as quantum wells, wires and dots, trions gain a sizeable binding energy and oscillator strength. They can be observed as distinct low-energy satellites to excitonic peaks in absorption or photoluminescence spectra. Quatrons are exotic four-particle complexes with an electric charge of  $\pm 2e$  that can occur in a variety of contexts, typically as a doubly charged exciton in a quantum dot. As an example of a particular specie of quatrons, the *Suris tetron* appears at intermediate concentration ranges in the two-dimensional electron-gas between the exciton-trion dominated regime at lower densities and the Fermi-sea at higher ones, with the binding of a hole to a trion, as identified by Koudinov et al. (2014). They can also be understood as trions repulsing the electron gas.

### Excitons in heterostructures

Excitons are even more important in systems of reduced dimensionality. These are possible thanks to the concept of heterostructure, that arose from the idea of forming an artificial lattice by alternating layers of two (or more) materials. Initially, this was to improve mechanical properties but the idea proved to be even more fruitful for electronic properties. In the limiting cases of a few layers, such “superlattices” result in a quantum confinement of the carriers and thus, in the case of semiconductors, of the excitons. This opens fascinating chapters of exciton physics in systems of reduced dimensionality, from 2D-excitons in Quantum Wells (QWs) to completely frozen excitons in Quantum Dots (QDs), passing by 1D-excitons in Quantum Wires. For a long time, such semiconductor heterostructures were mainly obtained by epitaxial growth. A more recent but increasingly popular type of heterostructures is that assembled by stacking layers of different 2D crystals. These are known as van der Waals heterostructures, due to the weak interaction between different layers (as opposed to the strong covalent bonds within each 2D layer). This major and fruitful development allowed by a breakthrough in the material physics of semiconductor 2D crystals, revisits the discipline as a whole by allowing an hitherto impossible atomic-level accuracy in the design of crystals, with no constraints from lattice mismatch at the interface. At the crudest level, depositing two flakes of such crystals realizes, by viscoelastic stamping, such a heterostructure. More complex structures are possible, in particular tri-layers, with a large-bandgap material interposed between two active monolayers to further control the recombination rate and dipole moment. Reduced dimensionalities bring a wealth of consequences on the physics of the respective excitons, starting with their density of states. Another immediate and illustrative consequence is the quantum confined Stark effect: applying an electric field perpendicularly to the direction of the exciton confinement does not result in its ionization, as is the case in bulk, since the heterostructure provides a safeguarding confining potential, but modifies instead greatly the exciton’s energy and oscillator strength. This increases drastically the radiative lifetime and produces giant Stark shifts of

the photoluminescence lines. This is due to the electron and hole being pushed in opposite directions by the electric field and thus reducing the overlap of their respective wavefunctions, which quantifies their recombination rate. Similarly to bulk excitons, quantum wire and quantum well excitons feature an energy continuum at the end of their quantized energy spectrum, due to their possible motion through the crystal. QDs are particular in that, by confining all the degrees of freedom, they effectively turn the exciton into a two-level system. Excitons are more relevant in optical QDs, since other types of quantum dots, formed by electrostatic gates, are better described in terms of uncorrelated electronic charges. The physics of excitons in various dimensions is different enough as to be treated independently, which we do in the next sections.

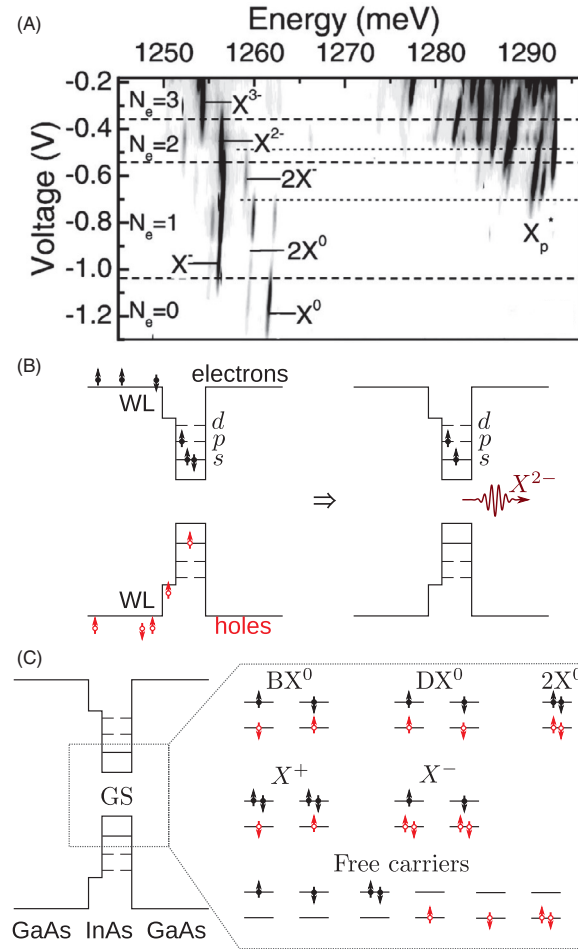
### Quantum wells, wires and rings excitons

Excitons emit photons within a radiative zone defined by the conservation of the in-plane or in-wire wavevector between the exciton and the photon, which is defined by a light-cone. Excitons with a larger momentum and thus outside of the light-cone, can be scattered there, by defects, phonons or other excitons, and thus act as a long-lived (non-radiative) reservoir. They also typically act as a background dynamical and spatial potential for the bright excitons. The radiative decay inside the light-cone is very fast, as shown by [Deveaud et al. \(1991\)](#). This led to a thriving field of ultrafast optics in the form of time-resolved photoluminescence. The radiative lifetime is determined by the exciton oscillator strength, and should produce a Lorentzian line (from its exponential decay in time). Other possible decay channels, in particular non-radiative, broaden but otherwise maintain this shape for the spectral line, in which case one speaks of homogeneous broadening. Pure dephasing enters in this category. Other processes can cause an inhomogeneous broadening that results from the emission of excitons of different frequencies, as is typically the case in presence of disorder due to fluctuations of energies in the crystal. This produces, instead, Gaussian lineshapes. Bright trions have different recombination rules than excitons regarding their momentum, since trion recombination leaves the third particle (electron or hole) in the crystal which can absorb the momentum leftover. This process, that still decays exponentially with increasing momentum, produces a characteristic low-energy tail in the trion spectrum.

An auxiliary between 1D and 0D confinement of the exciton can be found with so-called quantum rings (QRs), which close the loop of a wire so as to realize a unique geometry, specific to the solid-state. This makes them appealing for a wide range of problems related to rotational symmetry, discussed by [Fomin \(2014\)](#). QRs attracted special attention in the context of the fundamental problem of the geometric phase, also known as the Berry phase. They are particularly suited for the so-called Aharonov-Bohm effect that describes the geometric phase of a charged particle that goes both ways in a ring bathed in a magnetic field and that interferes with itself when completing the loop. Observable effects from a vector potential that produces no magnetic field are one of the most uncanny features of quantum mechanics and a spectacular manifestation of gauge theory. Excitons are neutral and the effect, first demonstrated with a charged exciton in a ring by [Bayer et al. \(2003\)](#), was thought to be too small for uncharged excitons, which indeed did not evidence it. However, the underlying electron and hole's geometric phases do result in a rich phenomenology for their wavefunction, that does produce an excitonic (also called optical) Aharonov-Bohm effect. This was first noted by [Chaplik \(1995\)](#) as peculiar properties of excitons confined in rings, with a theoretical dependence of their binding energy, effective mass and dispersion on a perpendicularly applied magnetic field. Those have been observed under an applied magnetic flux, first by [Ribeiro et al. \(2004\)](#) as oscillations in PL observables, in so-called type II QDs where the electron is bound by the dot but the hole repelled so as to produce a ring geometry (unlike in type I where both carriers are bound inside the dot), and then, in actual rings, as an oscillator strength periodically changing in time also detected in photoluminescence, by [Teodoro et al. \(2010\)](#).

### Quantum Dots excitons

Quantum Dots (QDs) are the ultimate confining crystallite, trapping particles in all three dimensions in systems with sizes ranging from sub-nanometers to small fractions of micrometers and made up from a few thousands only to millions of atomic nuclei. The first quantum dots were reported by [Ekimov and Onushchenko \(1981\)](#) who synthesized nanocrystals of copper chloride in glass, and observed a shift of their excitonic spectral line as a function of the heat-treatment to prepare the crystals, which they interpreted as an effect of size quantization. This was confirmed by [Éfros and Éfros \(1982\)](#) who described the nanocrystallite as a spherical box of varying radius for both one dot and an inhomogeneous ensemble. They also considered various limits of the size, leading to a separate confinement of the electron and hole or of the exciton as a whole. [Éfros \(2022\)](#) provides a detailed account of excitons in nanocrystals in this volume. Several ways have since been found to produce a broad family of QDs. For instance, molecular beam epitaxy can deposit a thin layer of, say, InAs on a GaAs substrate. The strain caused by the lattice constants mismatch lead to the spontaneous and self-assembled formation of little islands of various sizes and shapes depending on the growth conditions. By tailoring the shape, size and composition of the QD, one can tune the excitonic energy spectrum, its interaction energies, Coulomb energies, kinetic energies, electron-hole separations, oscillator strengths, etc. Typically, single QDs from an assembly each present a unique excitation spectrum. If dealing with such a group at large, the specificities of each individual are blurred in the averaging which results in broad emission spectra. Techniques have been developed to isolate single QDs, such as single-dot spectroscopy. This makes QDs rich laboratories to control and study electronic excitations. Such investigations led to describe them as "artificial atoms," in particular for their adherence to the basic atomistic rules of thumb, namely, the "aufbau" (filling-up of successive shells), and Hund's rule (occupying different orbitals of the same energy singly, before alternating spins). These account for how successive excitations get distributed in the energy levels of a QD, as shown in [Fig. 4A](#). When the system fills up with excitons—which is the case under optical excitation—one can in this way witness the successive formation of excitonic atoms



**Fig. 4** (A) Charging of multi-exciton states in a single InGaAs Quantum Dot, by [Finley et al. \(2001\)](#). The discrete spectral lines result from excitonic recombinations of states with a well-defined number of electrons and holes, such as  $X^0$  (exciton),  $2X^0$  (biexciton),  $X^{n-}$  (exciton with  $n$  electrons). The charging is here by lowering the applied voltage, which reduces the electrostatic potential of the QD to allow increasing numbers of electrons to tunnel inside (with corresponding thresholds indicated by the dashed horizontal lines), thus not allowing  $X^{n+}$  states with  $n$  holes, which are also possible in other configurations. The  $X_p^+$  lines at higher energies originate from  $p$ -shell transitions. (B) Example of a recombination responsible for the  $X^{2-}$  line: while the exciton recombines in the  $s$ -shell, it involves electrons in the  $p$  shells, (C) In the  $s$ -shell alone, in addition to the Ground State (GS), there are 15 possible configurations of excited states, with neutral Bright ( $BX^0$ ), Dark ( $DX^0$ ) and bi- ( $2X^0$ ) excitons along with their charged  $X^\pm$  versions or isolated electrons and holes. Panel (A) is reproduced with permission from Finley JJ, Fry PW, Ashmore AD, Lemaître A, Tartakovskii AL, Oulton R, Mowbray DJ, Skolnick MS, Hopkinson M, Buckle PD, and Maksym PA (2001) Observation of multicharged excitons and biexcitons in a single InGaAs quantum dot. *Physical Review B* 63:161305(R).

similar to hydrogen (a single exciton in the  $s$  shell), helium (two excitons in the  $s$ -shell or  $s^2$  in atomic notations), lithium (two excitons in the  $s$ -shell, one in the  $p$  shell,  $s^2p$ ), beryllium ( $s^2p^2$ ), boron ( $s^2p^3$ ) and carbon ( $s^2p^4$ ), as described and reported by [Hawrylak \(1999\)](#) and [Bayer et al. \(2000\)](#). When the system fills up with single electrons (or holes)—which is the case under gating or by doping—the single exciton remains the fundamental excitation but gets charged with subsequent excitations and one thus encounters trions  $X^{1\pm}$ , quatrons  $X^{2\pm}$  and higher-still excited states  $X^{k\pm}$  with, in the case of [Warburton et al. \(2000\)](#),  $k$  up to 5 when completing the  $p$  shell. These are referred to collectively as “excitonic complexes” or “multiexcitons.” The respective lines can be identified by a series of signatures, including the order in which they appear and disappear as the QD fills up according to Hund’s rules, the dependence of their intensity on excitation power, e.g., quadratic for biexcitons, their polarization, that is ruled by their spin composition and still other tricks, such as the lack of a fine-structure for the  $X^{1-}$  charged exciton that voids its exchange interaction or its large extent in gate voltage. Circular polarizations originate from exciton recombinations while linear polarizations from their quantum superpositions, which can be caused by a fine structure splitting. On the other hand, their distribution in energy indicates, e.g., whether electron-hole attraction or electron-electron repulsion dominates in the multi-exciton configuration, which can change depending on the lateral spatial extension of the electron and hole wavefunction, as shown by [Finley et al. \(2001\)](#) (cf. [Fig. 4](#)). Quantum Dots thus represent flexible artificial atoms, whose properties, although reminiscent of real atoms, can greatly vary in both qualitative and quantitative ways. For instance, the number of  $p$  and higher ( $d$ , etc.) shells thus becomes restricted to 2 in case of in-plane confinement for QD with cylindrical symmetry, as opposed to 3, 5, etc., for real (3D) atoms. There is also typically an asymmetry of QDs formed by interface fluctuations, which results in elongated shapes which further lift the degeneracy

of these states, resulting in a fine-structure splitting between them with polarized emission (those grown by colloidal chemistry, on the other hand, have a much greater spherical symmetry). Quantum dots are nowadays highly controlled both experimentally and theoretically. The fine-structure of their excitons is particularly well understood, with Bayer et al. (2002) taking into account spin interactions, such as the exchange Coulomb interaction, as well as hyperfine interactions with the nuclear spin from host atoms in the lattice, and the Zeeman interaction with an applied magnetic field. Rabi oscillations of QDs excitons have been first observed by Stievater et al. (2001) and demonstrated the validity of single excitons to realize the quantum optical physics of atoms, with  $\pi$ -optical pulse that revert their state, i.e., preparing the exciton in the one-particle state or, if already present, returning the system to the crystal vacuum by stimulated emission. The Rabi flopping could later be observed by Ramsay et al. (2010) for over 14  $\pi$ -pulses with a dephasing due to LA phonons and with a renormalization of the Rabi frequency attributed to fluctuations in the phonon bath, making the effect a solid-state analogue of the QED Lamb-shift for the hydrogen atom in vacuum. Related quantum optical milestones, such as the Mollow triplet of resonance fluorescence—where the exciton forms a quantum superposition with the laser photons to produce a ladder of equally-spaced dressed states—have also been unambiguously reported, for instance by Flagg et al. (2009).

### Direct and indirect excitons

Heterostructures allow to design complex arrangements of the confining potentials which permit to design sophisticated trapping for the electrons and holes, giving rise to new types of excitons, for instance by separating spatially their electron and hole. One can thereby grow two QWs (a so-called double QW) side to side and apply an electric field normal to their plane, forcing the electron in one well and the hole in the other (cf. Fig. 1C). Their binding across the barrier that separates the two wells leads to a spatially “Indirect Exciton,” denoted as IX. Same-well excitons are still possible and become known, in contrast, as spatially *Direct Excitons* (DX). The qualification of “spatial” is added if there is the need to distinguish from excitons in direct and indirect bandgap materials, where the separation is in momentum instead of in real space, and with the same consequence of decoupling the excitons from direct photon recombination/excitation, requiring, e.g., phonons to assist in the process (Fig. 1B). Recombination of IX is also possible, by tunnelling through the barrier, but is strongly reduced as compared to the DX one, as indeed the wavefunctions are spatially separated and their overlap becomes much smaller. This increases considerably their lifetime, by several orders of magnitudes in GaAs structures which can thus span over ns to tens of  $\mu$ s as compared to few ps only for direct excitons. Another advantage of IX is their strong-dipole moment  $ed$  aligned perpendicularly to the QW, with  $d$  the distance between the electron and hole layers (for this reason, they are also called “dipolar excitons”). Applying an electric field perpendicular to the QW plane results in an IX energy shift, known as the quantum confined Stark effect, and varying this field in space allows to create highly-tunable potential landscapes, of various shapes and extent, that can be static or dynamic. A diamond-shaped trap, for instance, allows to create a parabolic potential, which collects excitons and trap them at the center. One can thus apply a force through an electric field for these otherwise neutral particles, through a gradient of the field that turns into a gradient of energy shifts for the indirect excitons. Strong dipole moments also result in dipolar interactions that are stronger than the exchange interaction of direct excitons, and also of a long-range character  $\propto 1/r^3$  at large separations. Their repulsive character of oriented electric dipoles make them more stable against bound states or condensed phases. Indeed, the dipole polarization of excitons is not random but is imposed by the structure geometry and dipole-dipole repulsion prevents the formation of biexcitons. The same idea also applies to excitons in van der Waals heterostructures, as demonstrated by Rivera et al. (2018) (cf. Fig. 1D), with the advantage of a much enlarged space of parameters and with better control of the crystals. In that case, although the concepts and consequences are the same, one commonly speaks of “interlayer exciton” (ILX) instead of indirect ones. Besides, in addition of being spatially indirect, ILX can also be momentum-indirect, with the electron located in the  $K$  valley while the hole is located at the  $T$  point, as shown by Kunstmann et al. (2018). Quantum dots, as artificial atoms, can in a similar way be designed to form quantum dot molecules (QDMs), for instance by stacking two quantum dots on top of each other, as done by Schedelbeck et al. (1997). The exciton can now be formed by having its electron in one QD and its hole in the other, also producing an indirect exciton which, in a QD, becomes a solid-state counterpart of the  $H_2^+$  hydrogen molecule with one electron orbiting two protons, with the formation of bonding and antibonding states from the coherent excitonic wavefunction delocalized over the two QDs. The photoluminescence of QDM is markedly different from its single (atomic) QD counterpart: in addition to single lines indicating the multi-excitonic states, a rich phenomenology of crossing and anticrossing patterns emerge, involving states with much larger electric field dependencies thereby producing lines with very different slopes in the PL spectra as a function of the applied electric field. These are due to the strong Stark shift enabled by indirect excitons. The coupling of excitons and biexcitons, both inter and intra QDs, give rise to a characteristic X patterns, studied by Stinaff et al. (2006) and Scheibner et al. (2007), that are observed when the initial and final states undergo level resonances at similar electric fields.

### Key issues and trending topics

#### Rydberg excitons

Rydberg atoms are atoms whose outermost electron is excited in a quantum state  $|nlm\rangle$  with a high principal quantum number  $n$ . Since the effective size of the atom grows with  $n$  as  $\frac{1}{2}a_B(3n^2 - l(l+1))$ , Rydberg atoms are huge atoms with a correspondingly large dipole moment and thus a strong response to external fields as well as strong interactions. This makes them valuable for a wealth of

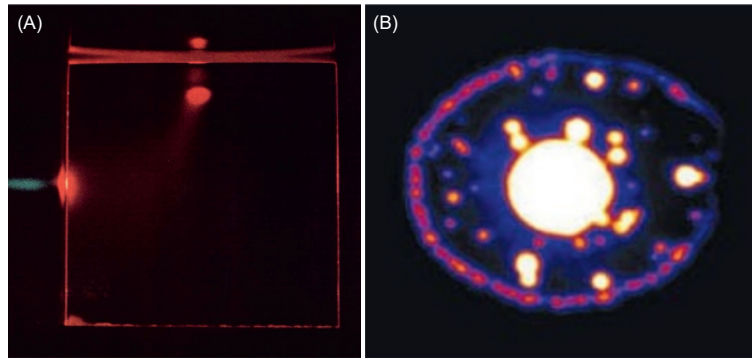
applications, such as their minute control in quantum optical experiments (leading to the 2012 Nobel prize, for “measuring and manipulation of individual quantum systems”). For the Rydberg exciton counterpart, a large binding energy is required, so that high values of  $n$  can be reached. From thermal dissociation alone, since the binding energy of the  $n$  shell scales like  $n^{-2}$ , the highest possible  $n$  is of the order of  $\sqrt{\frac{E_B}{k_B T}}$  which is about 6 for GaAs while it is close to 30 for  $\text{Cu}_2\text{O}$ , which is the material where Rydberg excitons are currently flourishing. Bringing excitons into a Rydberg state, as demonstrated by Kazimierzczuk et al. (2014) and shown in Fig. 2, is another compelling demonstration of their status of solid-state atoms. Low-level Wannier excitons being already large, their Rydberg version becomes gigantic, extending to Bohr radii as large as several micrometers, that is to say, over billions of sites of the lattice, for what remains a two-particle wavefunction. Despite stressing the excitonic picture of a bound state inside its host crystal to the limit, observations adhere remarkably well to relatively simple theoretical modellings. For instance, deviations from a pure Coulomb potential (such as is caused by the screening from inner electrons) can be taken into account through a so-called quantum defect  $\delta$  that renormalizes the hydrogenoid spectrum as  $E_n = -R_y/(n - \delta)^2$ . Rydberg excitons allow a detailed study of fundamental atomic effects, in particular of scaling properties: not only the size of the state but other characteristic properties scale with power-law dependencies on the principal quantum number, as studied in details by Heckötter et al. (2017). These include the binding energy (like  $n^{-2}$ ), the linewidth and the oscillator strength ( $n^{-3}$ ), leading to ultra-sharp resonances, and still other observables, such as the van der Waals interactions, scaling as  $n^{11}$  and thus producing a huge nonlinear absorption, referred to as Rydberg blockade, as per the blockade mechanism whereby the presence of one excited state prohibits the excitation of other states by direct repulsion. Furthermore, although of mesoscopic sizes and in a highly excited state, Rydberg excitons are believed to retain their quantum coherence, as was discussed by Grünwald et al. (2016). Rydberg excitons are therefore currently at one forefront of research into quantum-optical and exotic condensed-matter effects and could enjoy in the solid state the same fruitful path as their vacuum counterparts in a breadth of applications.

### Excitons in quantum Hall bilayers

An exotic breed of excitons consists in pairing, not an electron with a hole, but two electrons, in so-called electron bilayers, that are similar to the coupled QWs used for IX but such that both wells have conduction electrons and using a magnetic field instead of an electric one. This allows to bring the physics of Landau levels: the quantization of the electron cyclotron orbits under the applied magnetic field produces equally-spaced energy levels (the Landau levels) which are degenerate with a number of electrons per level proportional to the strength of the magnetic field. Each layer in isolation at half-filling of the lowest Landau level, constitutes a compressible Fermi liquid that behaves as a nonquantized Hall phase. Their pairing gives rise to new physics. Similarly to missing electrons in the valence band being treated as hole quasi-particles, an empty state for the Landau levels becomes an anti-particle with opposite charge. The fraction of occupied Landau levels, known as the filling factor, is tunable with the magnetic field. It is, in particular, possible to equate the number of electrons and holes, in which case one gets a counterpart of the standard “optical excitons.” Since such Landau levels are at the heart of the quantum Hall effect, the structures hosting such excitons are known as quantum Hall bilayers and the reliance on the magnetic field also brands their excitations as *magneto-excitons*. An immediate obvious advantage of the bilayer electrons, is that the excitons they form are stable: they do not recombine. They can therefore reach a true equilibrium with no race for thermalization against their finite lifetime, which is the main limitation for exciton thermodynamics.

### Excitonic matter

Once one recognizes that the excitonic picture of an effective hydrogen-like quasi-particle wandering freely within the crystal is valid, or at least useful, the next step is to keep on pushing the analogy and inquire whether condensed phases of excitons are possible, even though these would arise from quasi-particles of a more fundamental condensed system in the first place. The meta-analogy will be complete when one reaches the exciton state of excitonic matter itself. Interestingly, such questions have been entertained from the earliest days of excitonic physics, but remain among the most trendy and fundamental ones to this day. We now survey the most popular excitonic phases, starting with classical ones. The simplest is the gaseous phase, in the most extreme case of which excitons are assumed to be independent from each others, which, if at all possible, is to be sought at low densities. By sending laser pulses on a crystal and making snapshot of its luminescence (from exciton recombination) at different intervals, it has been possible to evidence clearly the drift of an exciton gas, accordingly to the kinetic theory of (normal) gases, in particular with the correct distribution of their velocities, with a temperature given by the lattice and a very large diffusion coefficient as mandated by the low exciton mass. Low-density excitons therefore qualify as a gas of particles confined inside the solid. Their mobility is considerably higher than even other solid-state particles, like electrons. Excitons have been shown by Tamor and Wolfe (1980) to drift under applied strain at speeds exceeding several hundred meters per seconds (as opposed to a few millimeters per seconds for an electron) over distances of several millimeters (cf. Fig. 5A). The strain is applied mechanically and causes a change of the bandgap, in turn resulting in a force applied to the exciton, which can be tracked by photoluminescence. To evolve from the dilute gas picture of excitons to more strongly correlated phases, one can simply increase the optical power to create more excitons. Doing so, one should not reach densities so high that excitons ionize into an electron and hole plasma, a process known as the Mott (1968) transition. Indeed distinct from a condensed phase of excitons, electron-hole liquids have long been known to exist but they arise when excitons themselves have been destroyed, or ionized, and thus, although interesting for their own sake, do not count as excitonic matter. A proper excitonic liquid can still be formed by the strong repulsive dipolar interaction between indirect excitons.



**Fig. 5** Two phases of excitonic matter: (A) drift of an exciton gas in  $\text{Cu}_2\text{O}$ , photographed by Trauernicht, Wolfe and Mysyrowicz (1984), showing how excitons created by a green laser (left) drift and recombine (red glow) in the minimum of a potential stress induced by pressure exerted on top of the 3D crystal, (B) At higher densities, indirect-excitons in 2D quantum wells, imaged by Butov et al. (2002) in-plane of the well, exhibit an unanticipated ring structure that is fragmented in a highly-ordered way. The laser that drives the system is in the central bright spot, or “inner ring.” The localized bright spots, which unlike the other rings do not follow the laser but remain fixed, are believed to be excitons trapped by defects. These images show how the excitonic picture, compelling in its gaseous phase, displays complex and original phenomenologies beyond expectations in its condensed phase. Panel (A) is reproduced with the permission from Wolfe JP and Mysyrowicz A (1984) Excitonic matter. *Scientific American* 250:98 and (B) from Butov LV, Gossard AC, and Chemla DS (2002) Macroscopically ordered state in an exciton system. *Nature* 418:751.

This is a regime of strong correlations, albeit believed to be of a classical character since it occurs with a negligible overlap of the excitonic wavefunctions. This phase is observed to behave thermodynamically, appearing abruptly below a critical temperature and above a critical density, and, when it is formed, to extend gradually to cover the full sample but with a low diffusivity. It also has a low conductance and its lineshape in PL is Gaussian and narrowing with increasing densities, like a liquid which is strongly correlated at short distances and as opposed to an uncorrelated gas which has a low energy tail characteristic of a fluctuating volume around the excitons. Its coexistence with another phase that matches an electron-hole plasma, reported by Stern et al. (2014), gives a picture of excitonic droplets. In between these droplets, it is believed that one finds a rarefied excitonic gas. After the gaseous and liquid phases, comes the crystalline phase. One has been predicted by Suris (2016) based on a calculated lower free energy for a triangular lattice than for the gaseous state, but in 2D and with long-range repulsive interactions, which is, however, the case for IX. Exciton interactions differing in the two phases, the desublimation (gas to crystal transformation) is predicted to yield strong qualitative signatures, with a clamping of two lines of photoluminescence as the excitonic crystal grows. At higher densities, when the system undergoes its Mott transition, a BCS-like mechanism for excitons has also been theorized by Keldysh and Kopayev (1965), that would also lead to a solid phase. In indirect bandgap semimetals, i.e., with a gap in momentum rather than energy, a spatial symmetry breaking (as opposed to  $U(1)$  symmetry breaking in a superconductor) should exhibit perfect conductivity yet with no Meissner effect (expulsion of magnetic field), which has been described as an excitonic solid, the “excitonium,” being also an analogy of a Wigner crystal but for excitons rather than electrons. In all these cases, there is the question whether the phase is classical, with their strong correlations and ordering being alike to solids, or, given the nature of the quasi-particles undergoing the transition, quantum. What constitutes specifically or genuinely quantum effects in such excitonic phases is still highly debated, as is overviewed in the next section.

### Exciton BEC and superfluids

Macroscopic quantum phases realized by the solid state and exhibiting exotic effects are not disputed, thanks to their broad range of compelling manifestations. These include superconductivity, superfluidity and Bose-Einstein condensation. Whether quasi-particles themselves can directly undergo such quantum transitions is much more open to debate. Superconductivity gives rise to Cooper pairs, for instance, but whether the superconducting phase itself can be understood as a Bose-Einstein condensate (BEC) of Cooper pairs is not widely agreed upon. The relevance, accuracy or even correctness of such an understanding as applied to any type of quasi-particles leads to many discussions in the literature. Excitons have been among the most hotly disputed quasi-particles to take part in such arguments. This is despite the fact that their delocalized nature throughout the entire lattice makes them intrinsically ideal to exhibit long-range order quantum coherence. While such macroscopic coherence for a single excitation is typically not expected despite being foundational to the theoretical picture, it can in fact be observed as was demonstrated by Dubin et al. (2006) with a single-exciton state—as compared to a dense gas—exhibiting fringes and so coherence at both ends (10  $\mu\text{m}$  apart) of a conjugated polymer chain (that behaves like a quantum wire). Nevertheless, the most basic picture of quantum coherence relies on the coherent teaming of a large assembly of bosons. The Bose statistics of excitons, as well as their diluted hydrogen-like gaseous structure at densities  $n \lesssim 1/a_B^D$  in a  $D$ -dimensional system, thus make them prime candidates for the phenomenon of Bose condensation. Moskalenko (1962) and Blatt et al. (1962) had considered them as strong contenders much before the first experimental BEC demonstration, which was with cold rubidium atoms in 1995. Such a Bose condensation can be understood as a condensation in momentum space where, consequently, the system becomes delocalized in space into a

macroscopic wavefunction, as opposed to exciton droplets, which are classical condensates in real space. The small effective mass and large Bohr radius of excitons make their critical temperature for 2D BEC  $T_c = 2\pi\hbar^2 n / (mk_B)$  a few K for densities of the order of  $10 \times 10^{10} \text{ cm}^{-2}$ , i.e., well within reach. Strictly speaking, in 2D, not BEC but a related phase transition—the Berezinskii-Kosterlitz-Thouless (BKT) one—describes coherence, which cannot be of infinite extent as it is destroyed by fluctuations, but decays slowly (as a power-law) instead. This is however for ideal systems while a real sample, in addition to be of finite dimensions, presents spatial fluctuations that act as local potentials in which 2D condensation becomes possible. This can be understood most simply by assuming a single potential that quantizes at least the lowest energy levels, or elaborately by considering the density of states of a disordered system that smooths out the 2D ideal case of an abrupt jump, into a 3D-looking one with a smooth vanishing of the state density. Besides, given that excitons have a large spatial extension, the picture of pointlike bosons eventually breaks down at large enough densities and their underlying structure becomes significant, in which case an electron-hole Cooper pairing should occur instead, as hypothesized by Keldysh and Kozlov (1968). In any case, despite favorable conditions from basic estimates, there are many difficulties to achieve exciton condensation. Unlike atoms, the exciton temperature is not directly accessible by direct cooling, only that of the lattice is and excitons are created with vastly larger temperatures, especially at the high densities required for their condensation. Their short radiative lifetime also makes it difficult to reach an equilibrium before excitons have recombined. The ground state of  $\text{Cu}_2\text{O}$  being optically inactive, it was hoped to circumvent this difficulty, but excitonic Auger processes (two-exciton collision annihilating one and transferring excess energy to the other) turned out to have a detrimental density/temperature contribution. To avoid coherence induced by the driving laser and let the system spontaneously form coherence through a spontaneous symmetry breaking, one should also seek signatures under non-resonant excitation, ideally, an electric one. A difficulty in such a case where all types of excitons are allowed, is that dark indirect excitons have typically slightly lower energy than the bright ones, due to the exchange interaction, and so the ground state of the exciton fluid should be dark, thus not directly optically observable, as indeed reported by Cohen et al. (2016). Not least is that even if the phase is actually realized, its unambiguous identification is problematic since a realm of alternative unrelated phenomena can produce a similar phenomenology, e.g., the Peierls phase—a spontaneous crystal distortion driven by the electron-phonon interaction and unrelated to excitons—for Bose condensation or the phonon wind—a drift force imparted by non-equilibrium phonons that push the excitons—pausing for superfluidity of particles with an already high mobility. Indirect excitons overcome several of the main limitations, with an increased lifetime and stronger interactions, added to the relaxation of their momentum conservation perpendicular to the plane that couples them to a continuum and thus speeds up their relaxation as opposed to the one-to-one coupling in bulk. This allows them to reach less than a tenth of the theoretical critical temperature within their lifetime. There are various ways to study the macroscopic quantum coherence of such excitons. To excite them non-resonantly, one can either create an excitonic population with a pulse and let it relax in time, or use continuous excitation but observe them farther apart from the laser spot after they drifted or have been transported there, and cooled down to lattice temperature. In the second scheme, an unexpected ring-structure, centered around the laser spot, with no emission in between, has been observed by Butov et al. (2002) (cf. Fig. 5B) and Snoke et al. (2002). While not apparent in Fig. 5B where the intensity gets saturated by the laser, Butov et al. (2002) actually resolves two kinds of rings: an internal one, which is remote from the laser-excitation spot by a distance comparable with the exciton diffusion length, in addition to the external ring, whose radius grows with increasing pumping power or decreasing applied voltage. This external ring can thus be sent to distances commensurable with the millimeter, away from the excitation spot, validating it as a macroscopic effect. It also exhibits a macroscopic pattern formation, consisting of periodically-spaced exciton bright spots referred to as a “macroscopically ordered exciton state.” This structure is pinned to the excitation laser beam and moves coherently with it if the beam is displaced. The whole structure of rings and spots collapses with increasing temperature into a Gaussian spot around the laser while the macroscopic ordering of bright spots appears below a critical temperature. The interpretation of this phenomenology is as follows: at high-enough density, excitons are freely moving particles, with strong dipolar interactions allowing them to screen potential fluctuations from the sample. Excitons that are moving too fast are beyond the light cone of possible radiative emission and thus do not emit photons. As they cool down, their individual kinetic energy lowers and they become radiative. The interplay of this with the potential landscape of the laser spot explains the inner ring. The outer ring, on the other hand, has been explained by Butov et al. (2004) and Rapaport et al. (2004) as exciton generation at the interface of the electron-rich (n) and hole-rich (p) regions. More intriguing is the periodic ordering displayed by the excitons in the external ring observed by Butov’s group. The periodic pattern has been attributed by Levitov et al. (2005) to a Turing-type of instability as applied to a Bose gas undergoing stimulated scattering. This not only identifies the phenomenon as quantum, unlike the rings and bright spots which can be understood classically, but also shows how exciton condensation exhibits original and unforeseen phenomena not found in any other system. The coherence of these excitons, as well as those from the bright localized spot, was established by High et al. (2012) both through the classic measurement of their first-order correlation function  $g^{(1)}(\Delta r)$  as well as from the build-up of polarization, both furthermore occurring together. The degree of coherence is larger on the edges of the spots, suggesting that the ideal density for condensation is not the highest one. The coherence length extends over several microns, so order of magnitudes more than for the classical exciton gas. Such dislocations and their ordering are reminiscent of vortices in superfluids and superconducting phases, and indeed phase dislocations have also been evidenced. However, their connection to the ring itself as opposed to isolated vortices, suggests a more complex and original structure of the excitonic condensate than had been theorized. Shift-interferometry experiments that allow to discriminate between various types of phase dislocations have been used by Leonard et al. (2021) to further support Bose condensation of indirect excitons by evidencing a flow of excitonic matter waves from localized bright spots, producing a moiré effect consistent with the observations. In other structures, similar quantum phases have been claimed. In quantum Hall bilayers, for instance, where lifetime is not an issue and critical densities for condensation can be fine-tuned,

the possible tunnelling of an electron hopping from one layer to the other signals a collective state of excitons, in the form of an ultra-sharp peak in the tunnelling rate for a critical separation between the layers, as was reported by Spielman et al. (2000). Such a peak can be explained as a pairing of electrons and holes in an excitonic, i.e., correlated fashion. Another dramatic observation by Kellogg et al. (2004) is the collapse of the Hall voltage when excitons are formed, as their electrical neutrality does not result in a flow of charge in the structure even though electrons and “holes” (that are also electrons) in both layers are in motion. In absence of their correlations (due to unmatched filling factors or distances between the layers beyond the critical one), two opposite Hall voltage cancel together but exist in each layer. In presence of such correlations, no voltage at all is present. This proves a correlated excitonic motion, as opposed to one of uncorrelated electrons and holes that cancels on average. The same effect was reported with indirect excitons in GaAs by Nandi et al. (2012) where it was described as a perfect Coulomb drag with a flow in on layer correlating a counter-flow in the other thanks to the condensation, and a similar phenomenology was also found with van der Waals semiconductors (MoSe<sub>2</sub>-WSe<sub>2</sub>), where a hBN barrier was used to suppress radiative recombination from interlayer excitons in the two TMD layers. The observation of a critical threshold for electroluminescence depending only on the exciton density and with a large and narrow peak of its intensity for equal electron and hole densities, also supports an excitonic BEC, from coherent and correlated pairing of the excitons from a condensed phase. While 2D systems have been the focus for the search of excitonic condensation since the early attempts in Cu<sub>2</sub>O, it has also been claimed in the 3D indirect bandgap semimetal TiSe<sub>2</sub> by Kogar et al. (2017) with a dedicated momentum-resolved electron energy-loss spectroscopy technique. That allowed them to probe the collective excitations and evidence the Goldstone mode of the excitonic condensate (exciton sound) that thus distinguishes the BEC phase from the Peierls one by exhibiting electronic as opposed to phononic modes. In all these cases, while the evidence is very strong, it still awaits consensual and unambiguous recognition that a BEC of excitons is, not only present, but to which extent, with which characteristics and with what relationships to related quantum phases. The complications of a finite lifetime in a disordered environment have furthermore been theoretically anticipated by Glazov and Suris (2018) to bring other and more surprising conceptual departures from equilibrium systems, such as requiring a finite temperature below which BEC becomes impossible, in contrast to the usual picture that the lower the temperature, the better. Excitonic condensates are thus clearly complicated objects as compared to their atomic counterparts, in a way that the single exciton is not. Although it does not transport neither charge nor mass, exciton superfluidity is another appealing quantum phase. The spectrum of elementary excitations of the excitonic BEC is expected to be linear and thus to adhere to the Landau criterion for superfluidity, by opening a gap for the possible excitations of the system, that is thus resilient to small perturbations, although other models regard the excitonic BEC as insulating.

## Emerging trends and future directions

### Moiré excitons

A twist made possible with the emerging van der Waals structures is to play with their angular crystallographic alignment, namely, to rotate one plane on top of the other (cf. Fig. 1E), something not possible with conventional crystal growth. This produces moiré patterns that in turn produce periodic and large-scale potentials, i.e., superlattices, with a characteristic wavelength that can range from a few to hundred nanometers. The excitons trapped in these particular landscapes give rise to a new brand of excitons: the moiré excitons, e.g., as reported by Seyler et al. (2019) in MoSe<sub>2</sub>/WSe<sub>2</sub> heterobi-layers. Inter-layer excitons are highly sensitive to such an angle, with strong variations of their PL emission, due to electron and holes at different K points having a finite momentum. This makes the corresponding excitons dark with zero momentum but becoming radiative if their exciton has the opposite momentum. This can be set by the angle, which controls the intervalley momentum separation between layers. Moiré excitons also exhibit spatially modulated selection rules for optical transitions with excitons localized at different sites of the moiré lattice and in different valleys emitting different polarizations. Short-range moiré lattices, on the other hand, produce minibands with folding of intra- and inter-layer excitonic bands, resulting in an hybridization between the intra- and interlayer excitons. This was reported by Alexeev et al. (2019) with holes from MoSe<sub>2</sub> binding to a twist-dependent superposition of electron states in the adjacent layers. The moiré potentials can be sufficiently strong to further quantize the trapped excitons, leading to antibunching (time separation) of their photon emission. The variety of phenomena ruled by the angle between the layer is so considerable that a new field of “twistronics” has emerged.

### Topological excitons, insulators and superconductors

While BEC and superfluidity have been, and remain, greatly discussed for excitons over several decades, more exotic quantum phases have emerged recently or are gaining increasing attention, especially when linked to the recent developments brought in condensed-matter physics by topology. A central topological phase is the Mott-insulator, describing a crystal that should be a metal according to conventional band theory but which, due to strong Coulomb interactions, behaves as an insulator instead. The simplest model to account for this physics is given by the Bose-Hubbard Hamiltonian, that rules the interplay between on-site interactions of bosons in a lattice and the energy tunnelling between adjacent sites. Such a phase has been already demonstrated with ultracold atoms, and was recently realized with dipolar excitons, in both triangular-lattice of moiré excitons in van der Waals structures (by Tang et al., 2020) and square lattices nano-patterned by metallic electrodes deposited on GaAs bilayers (by Lagoin et al., 2022). The Mott insulator phase consists of a distribution of the excitons to occupy each lattice site with a fixed and identical number of particles. Such non-fluctuating particle numbers are typical of strongly quantum correlated phases, as they break the

normally-distributed fluctuations of most classical systems. In the GaAs case, for instance, two Mott phases with one and two excitons per site, all in the same Wannier state, have been evidenced through spectral observations echoing in the PL the ordering of excitons in the lattice. Another insulating phase, this time directly attributed to excitons—the excitonic insulator—arises when the exciton’s binding energy competes with the bandgap and is thus particularly relevant in semimetals. This phase is now believed to have been observed by Jia et al. (2022) for the first time, in  $\text{WTe}_2$ , and to account for another paradigmatic topological phase but for electrons, the quantum spin Hall effect, that locks their momentum to their spin and thus constrains their motion on the edge of the sample in one direction only (preventing back scattering at zero magnetic field). It has also been linked to exciton condensation by Sun et al. (2022). Realizing and controlling such topological phases is important for prospects of harnessing quantum coherent effects, including condensation, superfluidity and superconductivity, as well as for building quantum simulators, that implement in a physical platform a model Hamiltonian that is too awkward to be described theoretically or computationally, but that can be simulated by observing the actual system. This could target topical systems such as high- $T_c$  and  $d$ -wave superconductors in other platforms, supersolids, Luttinger liquids, Kitaev models, generalized Kane-Mele model with a topological flat band, interaction-driven Haldane insulator, quantum magnets, Majorana modes, etc. An advantage of excitons as compared to electronic systems, such as graphene, is that optical recombination allows a convenient and precise observation of the underlying physics. Even when excitons themselves do not play the predominant role in such quantum phases, they can be crucial auxiliaries to aid or witness other quasi-particles into their quantum transition, such as for the formation of Wigner crystals (crystal of electrons) where excitons feel the periodic potential from the electrons, that undergo a typical scattering (Umklapp) producing a telltale signature in the form of a resonance. This approach has been exploited by Smoleński et al. (2021).

Among the most topical effects, superconductivity, at first sight, appears to be not relevant for excitons since they are neutral particles. A trion, however, is not, and conventional charge-transport is thus possible in this way, as demonstrated by Sanvitto et al. (2001), while their Bose condensation in a superfluid regime would result in their superconductivity, which was first suggested by Lozovik and Yudson (1976). Indeed, excitonic BEC in bilayers allows a net flow of charge intra-layers while still producing no net electric flow inter-layers if they are of the same magnitude but opposite directions, a mechanism that can be interpreted as “counterflow superconductivity.” Related mechanisms are important for superconductivity of paired electrons, for instance in graphene, but put more emphasis on the underlying constituents than on the excitonic quasi-particle as a whole. Less forthright for exciton superconductivity is a proposal from Ginzburg (1968) and taken-up by Bardeen (1978) himself, the historical figure who understood how phonons mediate superconductivity by pairing electrons into Cooper pairs. Together with Allender et al. (1973), Bardeen considers a bilayer structure, where phonons are to be substituted by excitons, the so-called “exciton mechanism” (of Ginzburg). In Bardeen–Cooper–Schrieffer (BCS) theory, the critical temperature is of the type  $k_B T_c = \Theta e^{-1/g}$  with  $\Theta$  the Debye energy (cutoff energy for a maximum phonon energy) and  $g = \mathcal{N}(0)V$  the product of  $\mathcal{N}(0)$  the electron density at the Fermi level and  $V$  the electron-phonon interaction, which result in the low-temperatures observed in conventional superconductors. One can thus raise  $T_c$  by increasing  $\Theta$  and/or  $g$ , which remains marginal with phonons. Substituting for excitonic values, that can produce a much stronger electron-electron attraction even if the retardation effect characteristic of BCS superconductivity is reduced. Taking both competing effects into account, one finds  $\Theta/k_B \approx 10^3\text{--}10^4$  K and  $g \approx \frac{1}{5}\text{--}\frac{1}{2}$  resulting in critical temperatures of several hundred Kelvin, which pioneered the quest for high-temperature superconductors. At still another level of the same idea, one can mediate Cooper pairing not with excitons which, just as phonon in BCS, are virtual excitations mediating the interaction, but with real excitons present in the system as a condensate, in which case their virtual excitations become those mediating the interaction. Since forming an exciton BEC is difficult, this can be eased by involving polaritons in a microcavity, as proposed by Laussy et al. (2010) or driving the excitonic BEC with a laser, thereby realizing a light-induced superconductivity mediated by excitons, which has also been predicted to possibly reach room temperature. While such effects have not yet been observed, related strongly-correlated quantum phases mediated by light, such as ferromagnetism of excitons, have indeed been realized by Wang et al. (2022) with moiré excitons exhibiting a characteristic hysteresis loop in their magnetic response.

### Excitonic devices

Excitonic devices already exist. They will probably invade the market as both technology and theoretical understanding progress and the transition from proof-of-principle demonstrations to a working superior excitonic technology will be achieved. In zero-dimension, quantum dot excitons already power some of the best quantum light sources, both at the single-photon level and for LED or laser applications. For the display industry, they offer high color purity and ultra-high resolution from thin screens, with also excellent brightness and a large color gamut, including perfect black. For lasing, excitons in colloidal quantum dots—that already provide efficient photoluminescence over a wider spectrum than their epitaxial counterpart—have also demonstrated with Dang et al. (2012) single-exciton gain, allowing to implement highly-efficient, low-threshold full-color lasers. For quantum applications, multiplexed (i.e., synchronized) QDs have, among other services, already allowed to claim quantum supremacy in boson sampling. All these feasts follow from the highly-tunable excitonic properties in artificial atoms that can be molded at will in a thriving material physics. The QD exciton that behaves like a two-level system becomes, in quantum mechanical terms, a qubit, i.e., the fundamental unit of quantum information processing which can sustain superpositions of states and nonlocal entanglement, needed for quantum information processing. Coupled excitons in a single QD have been used in this way by Li et al. (2003) to build a two-qubit gate. In a paradigmatic which-way recombination, the biexciton’s decay to its ground state through a radiative cascade passing by one or the other exciton, leads to entanglement of the emitted photons, as shown by Akopian et al. (2006). By quantum-correlating deterministically hundreds of photons—so-called cluster states, able to power universal quantum

computation—the interplay of dark excitons with the biexciton through a precise timing by optical pulses allowed Schwartz et al. (2016) to entangle the polarization in a stream of emitted photons. The spin-degree of freedom is useful for that purpose, thanks to its long coherence time, due to a weak interaction with the environment, yet with a great ease of direct control with optical pulses. This allowed Kroutvar et al. (2004) to implement optically fully-programmable spin memories. In two-dimensions, there is the prospect of taking advantage of exciton propagation, and thereby to implement more powerful functionalities. This, however, comes at the price of being more difficult to master and only proof-of-principle demonstrations of 2D excitonic devices, including some with similar functionalities as those above (light-source, memory storage, etc.), have been demonstrated. Transport applications start with the exciton conveyors, such as those implemented by Winbow et al. (2011) and Peng et al. (2022), that literally transport excitons on an electrostatic carpet powered by surface acoustic waves, whose amplitude and speed are directly controlled through the applied voltage and AC frequency, respectively. A near-field piezoelectric field periodically modulates the exciton energy to drag them over distances an order of magnitude greater than by diffusion alone, that is already very large. This could develop into a new kind of excito-phononic circuits. A technological grail is the transistor, whose electronic implementation comes with the drawback of high energy consumption and heat dissipation. Purely photonic transistors lack the nonlinearities inherent to the device. Excitons thus appear an ideal compromise to realize an opto-electronic transistor, also interfacing the two basic tenets of technology: electronics (control) and optics (communication). Excitonic transistors as well as their combination in a circuit, have been demonstrated by High et al. (2008) to effectuate such a conversion of photons into excitons that flow through the device before their ultimate reconversion into photons. The principle of an excitonic transistor is, similarly to its electronic FET model, to control via an applied voltage the flow of excitons through two terminals conditioned to the state of a third one. A transistor acting as a switch (as opposed to as an amplifier, the other typical function of a transistor) was demonstrated, with a control of the flux of excitons in the source electrode by a gate voltage. At the heart of the mechanism, an applied electric field creates an energy barrier or trap for IX and therefore can suppress or enable exciton diffusion. From the optical side, the exciton recombination rate can thus be controlled with an applied electromagnetic field. Three such excitonic transistors have also been coupled to share a common electrode, thereby realizing an excitonic circuit, with one application implemented to directionally switch the exciton flux. At this time, 2D excitonic transistors are not functional enough to power a substituting technology. The binding energies of the materials where they have been implemented require cryogenic cooling, and their performances do not yet compete with even decades-old electronics. While quantum devices would be worth running at cryogenic temperatures, classical devices should certainly operate at room temperature to rival those already existing, in which case strongly-bound excitons such as those provided by van der Waals structures are required. Emerging materials could, indeed, push forward excitons as strong contenders to replace electronics. These also reinforce the role of the excitons over the electrons and holes in conventional semiconductor devices, due to their strong binding, oscillator strengths and Coulomb interactions, so that new generations of excitonic LEDs, junctions, electrically controllable routers and transistors become possible, operating with currents as low as nanoamps. In the case of the transistor, room-temperature operation with a transport of excitons over five micrometers distance, has already been demonstrated by Unuchek et al. (2018). In addition to that, the rich valley-physics opens new functionalities, such as the control of light handedness that allowed Zhang et al. (2014b) to implement flexible chiral optoelectronics. These rapid developments open the route, not only toward an excitonic computer, but also entirely new types of devices such as transparent and stretchable screens.

## Conclusion

Excitons have been part of condensed-matter physics since its early days and they remain at the forefront of its most active, fruitful and promising developments. They have retained throughout their history a mixed nature of being a compelling quasi-particle and a dubious one to truly or rigorously account for the complexity of condensed matter systems. In both cases, they are central, in one form or another, to a vast and varied range of phenomena. Even as they get tentatively discarded from even their excitonic resonances, they re-appear in the most unexpected places, such as metals. Excitons will likely remain unavoidable mental pictures, forever introduced as electron-hole pair bound states, and as technological applications—possibly room-temperature superconducting excitonic quantum devices—develop from ideas based on their conception, they will continue to occupy a central position in condensed-matter physics.

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