Emerging collective quantum phenomena of excitons in metal-halide perovskites

Ajay K. Poonia, Parashurama Salunkhe, Angshuman Nag, and K.V. Adarsh*

Metal-halide perovskites (MHPs) with unique electronic and optical properties have emerged as promising materials with a broad spectrum of applications in photovoltaics, optoelectronic, and photonic devices. The distinct properties and tremendous potential of MHPs are intricately defined by excitons and collective quantum states. This article reviews the excitonic states and coordinated interplay of charge, spin, and lattice. We discuss the recent experimental and theoretical discoveries of excitonic phenomena, as well as correlated states involving condensation and cooperative emission. Additionally, our exploration extends to the structural properties of MHPs that facilitate the emergence of robust quantum states, even at room temperatures. Finally, an overview of the remaining challenges and potential applications of MHPs in quantum optics, coherent light sources, electrically driven amplified spontaneous emission, and superfluorescent lasing is provided.

Introduction

Metal-halide perovskites (MHPs) and their low-dimensional structures, with unique combinations of spin, charge, and lattice degree of freedom, have emerged as a revolutionary class of material for a range of technological applications,[1–8] as shown in **Figure 1**. Their extraordinary performance in photovoltaic devices, light-emitting diodes, lasing, sensing, detection, and quantum devices has propelled them to the forefront of materials science research.[1–4,9,10] The credit goes to their unique physical and chemical properties, such as defect tolerance, tunable bandgap from ultraviolet (UV) to near-infrared (NIR) regime, strong spin–orbit coupling, high photoluminescence (PL) quantum yield, strong electron–phonon interaction (EPI), inherently rich quasiparticle states, and a facile low-cost solution-based synthesis method that is potentially suitable for large-scale manufacturing.[1,4,11,12]

MHPs have been known since the 1890s, and they were studied for their optical properties even in the 1950s.[1] However, a turning point came in 2009 when they attracted the unprecedented attention of the scientific community after the reports of power-conversion efficiencies of 3.8% by Kojima et al.[13] Hybrid MHP solar cells crossed the power-conversion efficiency of more than 25% within a decade.[10,14] Soon, the MHPs got attention for diverse applications, including light-emitting devices, lasing, spintronics, neuromorphic, and quantum devices. The unique properties that drive this second wave are the near unity PL quantum yield, narrow linewidth of emission, single photon emission, clean spin selection rules, highly spin-polarized carriers, combined electronic and ionic conductivities, macroscopic quantum states, etc.[1–3,8,9]

There are many prior review articles covering the synthesis, structure, properties, and application of MHPs.[1–4,9,10] This article seeks to illuminate the emerging many-body collective phenomena of excitons in bulk and low-dimensional MHPs. First, we discuss the fundamental excitonic properties of MHPs that govern the light-absorbing and light-emitting devices. Subsequently, we will delve into the intricate play between excitons, polarons, and photons that give rise to the many-body states of exciton–polaron, biexciton, exciton–polariton, Floquet states, cooperative emission, and condensation. Finally, we will briefly discuss the prospects of emerging macroscopic states for quantum technologies.

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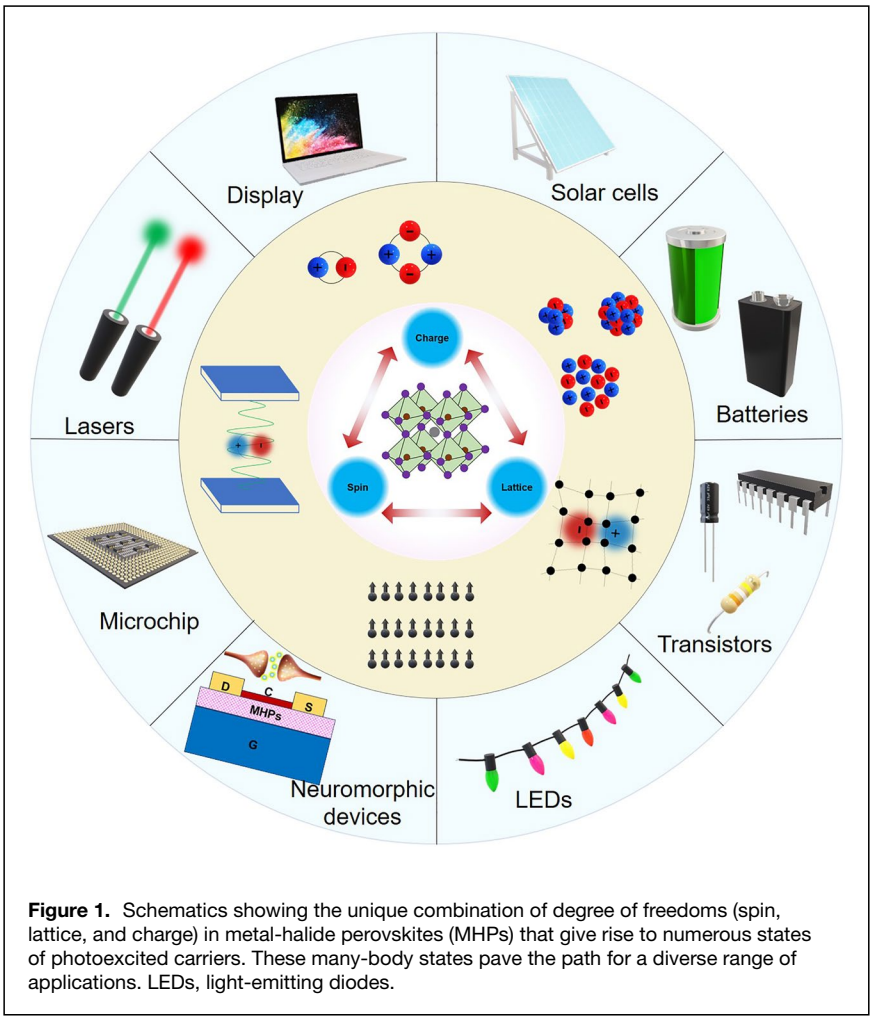

Figure 1. Schematics showing the unique combination of degree of freedoms (spin, lattice, and charge) in metal-halide perovskites (MHPs) that give rise to numerous states of photoexcited carriers. These many-body states pave the path for a diverse range of applications. LEDs, light-emitting diodes.

Excitons in MHPs

The photoexcitation of semiconductors creates electrons and holes in the conduction and valence band, respectively. These oppositely charged carriers interact via Coulomb interaction and lead to the formation of an electron–hole quasiparticle, known as an exciton. Excitons play a crucial role in the properties of semiconductors and in the designing of various devices, hence making them a topic of intense interest and research.

Typically, in the bulk MHPs, excitons are of Wannier–Mott type and characterized by low binding energy (<50 meV) due to strong dielectric screening.[15] Therefore, at room temperature, excitons coexist with free carriers in a dynamic equilibrium. The low exciton binding energy in bulk MHPs is key to their success in solar cells. The efficient absorption is balanced with the need for the exciton to reach an interface and dissociate into free charges before recombination. The energy spectrum of excitons in MHPs is also characterized by the study of excitonic Rydberg states,[16–18] where multiple internal transitions within Rydberg excitons are explored. The optically bright hydrogenic states (1s, 2s,….) and transition to the optically forbidden dark states (2*p*, 3*p*,….) were explored to characterize the formation, Bohr radius, and binding energy of excitons. When the crystal size of MHPs shrinks to a few nanometers or the crystal structure becomes low-dimensional such as layered hybrid perovskites, the excitons experience a drastic change due to quantum confinement. They display unique and enhanced properties, such as enhanced binding energy into the hundreds of millielectronvolt range, high PL quantum yield, sharp linewidth, and optical anisotropy.[2] The sizes and shape of the confinement in MHPnanostructure directly influence the excitons and offer tremendous potential for tuning their properties in a controlled way. For instance, the rate of thermalization, Auger recombination, and optical gain of excitons can be modulated by changing the shape and size of the nanostructures.[2,19]

A deeper understanding of excitons comes from breaking down the band-edge states and associated quantum numbers.[20] In the MHPs, the conduction band edge consists of Pb 6*p* orbitals, and strong spin–orbit coupling makes the bottom of the conduction band doubly degenerate, and the electrons can occupy $|\pm 1/2>$ spin states. The valence-band maximum consists of Pb 6*s* and halide atomic orbitals (Cl 3*p*, Br 4*p*, I 5*p*) with overall *s* symmetry, which leads the holes to be present in two spin states $|\pm 1/2>$. Thus, MHPs have a simple band-edge structure compared to III–V and II–VI semiconductors, where heavy and light hole subbands make the exciton transitions relatively complex.[21] Due to this, in MHPs, the optical transitions of excitons are governed by clean spin selection rules, opening up avenues for exotic properties. For instance, circularly polarized light can selectively excite electrons with a specific spin orientation into the conduction band, creating a population of spin-polarized carriers. This controllability not only makes MHPs an ideal platform for studying fundamental spin dynamics, but also facilitates investigations of many-body phenomena such as spin-selective biexcitons, bandgap renormalization, and ultrafast manipulation of electronic states via the optical Stark effect.[9,22–26] The inherent potential of spin-polarized excitons within MHPs paves the way for future device concepts in the emerging field of spintronics.[9]

One intriguing aspect of excitons in MHPs is their fine structure, which is influenced by several factors, including electron–hole exchange interaction, crystal symmetry, and potentially the Rashba effect.[15,27–29] As per orbital and

magnetic quantum number, initially, excitons possess four possible states [0 ($J_{ex}=0$) and $0,+1,-1$ ($J_{ex}=1$)]. The exchange interaction then splits these into an optically inactive dark singlet state and an optically active bright triplet state. Depending on the specific crystal structure (tetragonal, orthorhombic, or cubic), the bright triplet could further split into sublevels with distinct light polarization properties. Interestingly, in view of the unusually short PL lifetime observed in MHPs at low temperatures, a possible reversal of the typical bright and dark state ordering was suggested. While the exchange interaction alone does not seem sufficient to explain this, the Rashba effect has been proposed as an alternative mechanism.[15] However, this explanation is challenged by observations such as PL decay patterns, the temperature dependence of light emission, mixing of dark-bright states, and direct observation of the dark singlet ground state. More recent interpretations suggest that weak coupling to acoustic phonons and larger longitudinal optical (LO) phonon energies relative to the bright-dark splitting could be responsible.[15] Overall, understanding the precise nature of exciton fine structure in MHPs remains an active and debated area of research.

Many-body complexes of excitons

Exciton–polaron

Excitons and polarons are two quasiparticle states of photoexcited semiconductors, consisting of electron–hole pair and electron (hole)–phonon, respectively. They have their unique fingerprints and impact the electronic, optical, and transport properties of the semiconductors in a distinct way. For instance, the properties of low-polar semiconductors (such as GaAs, Si, and InAs) are generally governed by excitons. Meanwhile, in highly polar semiconductors (such as $SrTiO_3$), polarons determine the properties of the system. Interestingly, the properties of MHPs cannot be described in terms of separate exciton and polaron framework; but they are deeply intertwined and dually affect the carrier dynamics.[11,30,31] Even though excitons are charge-neutral particles and a weak EPI is expected; however, the local spatial fluctuations in the charge densities of electrons and holes can induce the lattice distortions.[32] This leads to a complex quasiparticle called an exciton–polaron. It can be thought of as an exciton dressed in a cloud of lattice vibrations.

MHPs are characterized by relatively soft crystal lattices that result in strong interactions between electrons (or holes) and the lattice vibrations (phonons).[11] The resulting polaron formation determines the effective masses of the carriers in the spin split bands, where phonon dressing is decided by the Fröhlich coupling strengths (α)[33]

$$\alpha = \frac{e^2}{4\pi \in \hbar}\left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0}\right)\sqrt{\frac{m^*}{2E_{LO}}}. \quad (1)$$

Here ϵ, ε_∞, ε_0, m^*, and E_{LO} are dielectric constant of vacuum, optical dielectric constant, static dielectric constant of the medium, effective mass of carriers, and energy of LO phonon. The larger value of α in MHPs compared to other inorganic semiconductors[15,34] (such as CdS, GaAs, SiC) makes them a favorable ground for the polarons. In such a situation, it is important to consider the polaronic nature of excitons to fully describe the carrier dynamics of MHPs. The polaronic nature of excitons has been found to play crucial in the MHPs by affecting the binding energy, optical, electronic, and transport properties.[15,22,24,35–39] For a better understanding of the complex exciton–polaron dynamic, the 3D hydrogen atom model with the modified potentials has also been incorporated, details of which can be found in Reference 15. Further, exciton–polarons are more important in 2D hybrid MHPs, where interaction between excitons and lattice vibrations is remarkably strong. The polaronic nature of excitons is also found to influence the inter and intra-Coulomb interaction, increasing exciton spin lifetime by reducing electron–hole wave-function overlap.[39,40]

Biexciton

At a high enough intensity of photoexcitation, the inter-exciton distance decreases, enhancing the exciton–exciton interaction. In the situation of attractive interaction, a bound state of two excitons is formed, which is termed a biexciton. In MHPs, exciton–exciton interactions have been studied using transient absorption, PL, and two-dimensional (2D) spectroscopy, suggesting biexciton formation with intermediate to high binding energy in the range from a few to more than 100 meV.[41–44] The biexcitons play a critical role in the emission and many-body effect of MHPs, showing the amplified spontaneous emission (ASE)[19,45–47] and optical Stark effect (OSE) (as discussed in detail in the later section) even at room temperature.[23,48]

Interestingly, akin to its impact on the exciton dynamics, EPI also plays a key role in the exciton–exciton interaction and can enhance the attractive interaction between two excitons to form a ground biexciton state.[24,35,49] Several studies suggest the idea of a polaronic biexciton[24,35] and proposed that in the MHPs, EPI induces the bipolaronic coupling of two same charge carriers that give rise to unconventional exciton–exciton attraction mediated by phonons.[24]

Exciton–polariton

Exciton–polariton is the hybrid light–matter quasiparticle that arises from the strong coupling of excitons and photons. They have properties of the exciton as well as of photons such as low effective mass and large nonlinearity. They are considered as an important many-body state for the development of low threshold lasing sources, Bose–Einstein condensation, logic devices, etc.[50,51]

A vital requirement for their manifestation is the presence of robust excitons. Consequently, MHPs have emerged as a choice for active material for the exciton–polariton.[50] Although the first study of exciton–polariton for MHPs dated back to 1998 using spin-coated film of $(C_6H_5C_2H_4NH_3)_2PbI_4$, it suffered from the low quality factor and low polariton lifetime.[52] The recent advance in MHP synthesis, processing, and photonic architecture lead to the effective polariton with

an increase in quality factor up to 2000 and Rabi splitting more than 500 meV.[50] In fact, in MHPs, exciton–polaritons have been realized even at room temperature, along with the Rydberg exciton–polaritons and their condensation.[50,53] Further, by using the perovskite zigzag lattice structure, exciton–polariton topological insulators were also demonstrated, and an ability to switch an exciton–polariton system between different topological phases using the polarization of light has been shown.[54]

Emerging phenomena

The unique ground of MHPs where excitons and polarons coexist simultaneously and interact strongly with photons offers an ideal platform for achieving the collective phenomena of excitons and new functionalities.[55–58] Further, the relatively soft and dynamic lattice of MHP lattice with strong-intermediate EPI is another key property of MHPs that makes it one of the good hosts for various quasiparticles and their many-body effects.[59] There are plenty of noteworthy emerging phenomena that have been explored in MHPs; here, we will focus on (1) collective and cooperative emission, (2) electron–hole liquid (EHL), (3) exciton–polariton condensation, and (4) Floquet states, which are discussed next.

Collective and cooperative emission

MHPs show ASE and superfluorescence (SF), which is also known as mirrorless lasing, offering a pathway to high-performance, low-cost, and tunable light sources. In a typical radiative decay, the excited dipoles or emitter relax through the spontaneous emission of incoherent photons. However, if one spontaneously emitted photon stimulates the emission of another photon, then optical amplification takes place. This cascade emission and amplification of the light take place as a collective effect, where the bunch of emitted photons have the same energy and direction, known as ASE.

Due to the large absorption coefficient and high optical gain, MHPs show extraordinary capabilities of low threshold ASE.[60] Importantly, the composition of MHPs can be meticulously adjusted to engineer their bandgap and, consequently, tune the wavelength of ASE across a broad range of spectrum, as shown in **Figure 2**a. Further, nanostructures and layered MHPs provide a unique framework for enhanced ASE through electronic confinement.[60,61] Although achieving the ASE in MHPs under pulse excitation was relatively facile, the true challenge is faced under continuous-wave (CW) excitation. By using the film of mixed cation and anion MHPs with CW excitation, ASE is achieved up to a temperature of 120 K,[62] as shown in Figure 2b. Further, truly electrically excited ASE is still missing, even though in this direction, progress has been made by getting the electrically assisted ASE up to a temperature of 77 K[63,64] (see Figure 2c).

On the other hand, if the radiative field of spontaneously emitted photons causes the ensemble of dipoles to couple with each other, then they form a macroscopic dipole, as shown in Figure 2d. In such conditions, the system releases the stored energy in the form of a coherent burst of light, which is known as SF.[65,66] With the ability of supporting collective action of carriers, the self-assembly of MHP nanocrystals[67,68] turns out as an ideal platform for SF. The pioneering study by Rainò et al. demonstrated SF within superlattices of MHP quantum dots at 6 K.[68] Key signatures of SF, including spectral redshifts shown in Figure 2e, accelerated radiative decay (Figure 2f) along with photon bunching and Burnham–Chiao ringing, provided compelling evidence for SF. The following studies by Biliroglu and Findik et al. in $MAPbI_3$ (MA: methylammonium) thin film expanded the potential temperature horizon for observing SF emission at temperatures beyond 78 K.[69] In a later study,[70] the same authors uncover the SF even at room temperature in quasi-2D PEA:$CsPbBr_3$ (PEA: phenethylammonium). Demonstrating room-temperature SF is a significant fundamental advancement, as it traditionally required extremely low temperatures. The room-temperature SF opens an avenue for developing a technology for a new generation of efficient light-emitting devices based on MHPs that operate at standard ambient conditions. As demonstrated in Figure 2g, SF at room temperature was uncovered in the form of an intense sharp emission above a threshold pump fluence, along with a strongly fluence dependency in PL lifetime(see Figure 2h). Importantly this study also put forward a mechanism for the protection of coherence in PEA:$CsPbBr_3$, and suggested that formation of exciton–polaron effectively shields the excitons for room-temperature SF.

Interestingly, in a recent study, SF was demonstrated in the highly disordered state of electron–hole plasma at temperatures as high as 175 K.[71] The low pump fluence regime consists of spontaneous emission and ASE, while SF emerges in the high pump fluence regime of an electron–hole plasma, as shown in Figure 2i. However, in the regime of electron–hole plasma, the polaron is unstable[69] and seeks a different mechanism for the protection of decoherence and the occurrence of stable SF from electron–hole plasma. One possible mechanism related to polaron formation is the EPI, which supports the formation of a quasi-thermal state of electron–hole plasma for the occurrence of SF.[34] By further pushing the boundaries of established SF and superradiance concepts, single photon superradiance behavior also demonstrated in individual cesium lead halide perovskite quantum dots.[72] While traditionally SF or superradiance requires cooperative behavior from many excited emitters, these intriguing findings point to the superradiance behavior for a single excitation. Such superradiance dynamics for a single excitation emerge due to wavefunction delocalization in a large size quantum dot (relative to exciton Bohr radius). This has further paved the way for potential advances of MHPs in single photon generation with tailored spectral and temporal properties for quantum optics applications.

Along with these experimental studies, the cooperative emission and exciton localization over quantum dots in MHPs are also studied theoretically.[73–75] The factors affecting the coherence

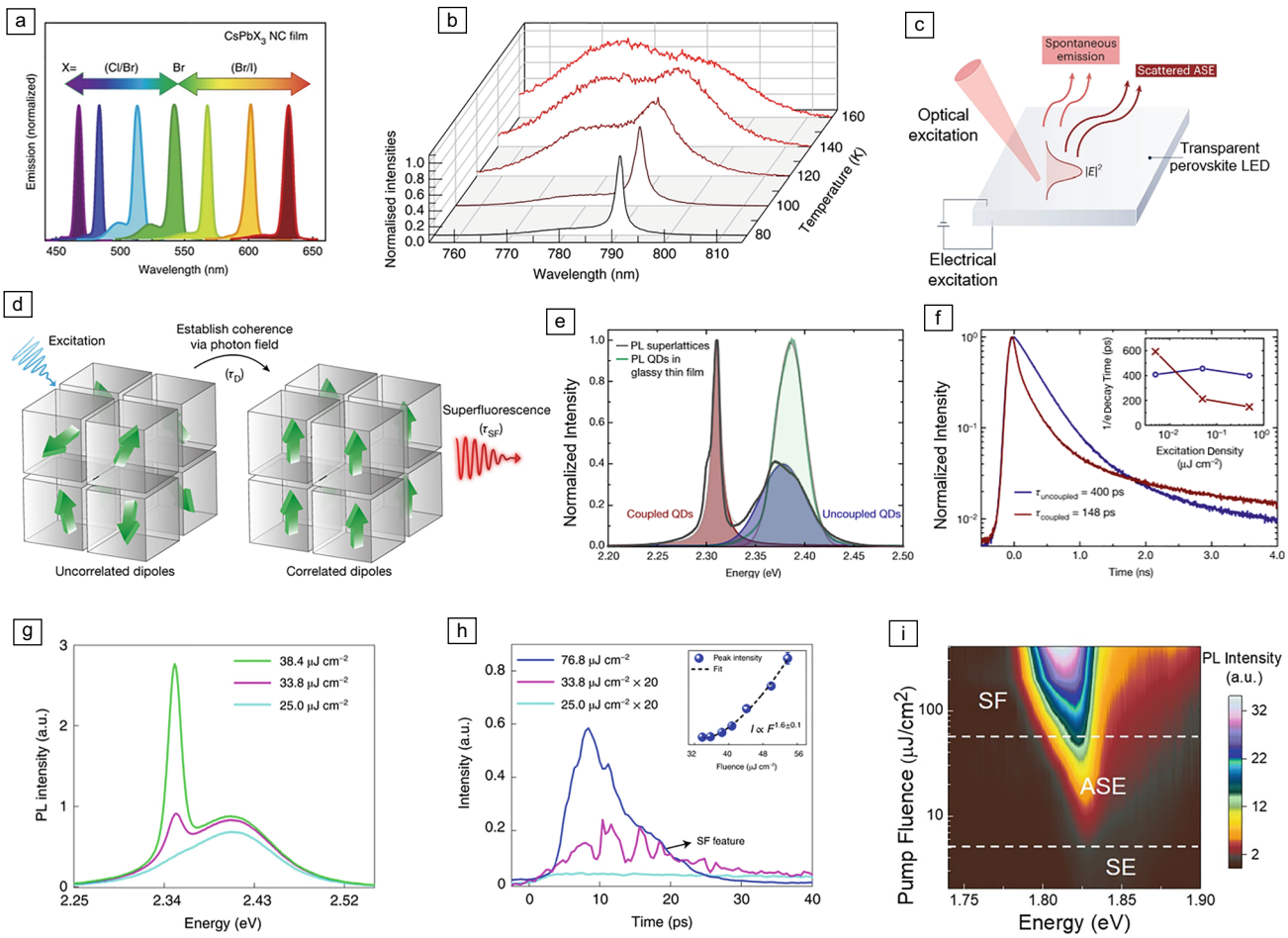

Figure 2. (a) Emission spectra of $CsPbX_3$ nanocrystal (NC) film, showing low threshold amplified spontaneous emission (ASE) across the entire visible regime.[60] (b) Temperature dependence emission of mixed metal-halide perovskites under continuous-wave excitation shows the ASE up to 120 K.[62] (c) Schematic diagram for getting electrically assisted ASE with both optical and electrical excitation.[64] (d) Schematic diagram showing the emergence of superfluorescence (SF) from the initially uncorrelated ensemble of dipoles.[68] (e) Photoluminescence (PL) spectra for different cases of coupled and uncoupled quantum dots (QDs) and glassy film of $CsPbBr_3$, distinguishing SF from spontaneous emission.[68] (f) PL lifetime showing rapid decay of SF.[68] (g) Excitation dependence PL spectra of quasi-2D $CsPbBr_3$ thin film at 300 K. SF emerge after threshold fluence of 33 μJ/cm^2.[70] (h) Time-resolved PL decay profile for spontaneous emission and SF.[70] (i) Color plot of excitation dependence PL of $CsPbBrI_2$, revealing spontaneous emission (SE), ASE in the low pump fluence regime of excitons, while SF from the high pump fluence regime of electron–hole plasma.[71] LED, light-emitting diode.

formation, the role of thermal effects, scattering with optical phonons, static disorder, and defects in decoherence were discussed. These studies revealed the major hindrances, which are inherent to the MHPs and restrict the SF, such as structural imperfections are highly disruptive to the delicate phase relationships required for long-range superradiance, particularly at room temperatures. Studies addressing temperature dependence highlighted the need for strategies to overcome the phenomenon of thermal decoherence. Further, it is demonstrated that adjusting factors such as nanocrystal shape, lattice dimensionality, and energy alignment hold the key to promoting longer-range coupling and bolstering superradiance against disorder.

While substantial progress has been made in observing SF across both MHP nanostructures and polycrystalline films, several questions remain unanswered. Unlike classic systems, SF in MHPs persists even at room temperatures and high carrier density regimes. It remains a key puzzle as to how these materials retain the delicate electronic coherence demanded by SF. Although superlattice formation provides advantages, evidence suggests additional mechanisms for the protection of coherence could play a crucial role. Factors such as strong EPI (acting as quantum shock absorbers and supporting quasi-thermal ground state) could reduce nonradiative losses and help to achieve quasi-equilibrium states supportive of SF processes.

Electron–hole liquid

The electron–hole liquid (EHL) is a striking manifestation of Fermi condensation in semiconductor materials when

they reach below a critical temperature and above a carrier density.[76,77] It represents a first-order phase transition of excitons, where these excitons dissociate into free carriers, leading to the formation of a liquid-like state consisting of electron–hole droplets as shown in **Figure 3**a. This phenomenon of gas (insulator) to liquid (metallic) transition is particularly intriguing due to its low mass particles, constant carrier density, and large optical gain, making it a subject of extensive investigation across various semiconductor materials.[78] Initially, due to the association of thermal effect with excitation, EHL is believed to be primarily stable in indirect semiconductors. The long lifetime and multivalley structure of the indirect bandgap provide enough time for quasi-thermal equilibrium. However, theoretical work by Beni and Rice[79] and Keldysh and Silin[80] suggested that direct bandgap polar semiconductors with enhanced EPI could stabilize the ground state of EHLs. In this way, MHPs, characterized by intermediate to strong EPI, have emerged as promising candidates for realizing EHLs at higher temperatures.

In single crystals of $CH_3NH_3PbBr_3$, EHL was observed at room temperature, evidenced by the emission's consistent position and optical hysteresis, as shown in Figure 3b–d. These features are characteristic signatures of EHL.[81] Investigations on films of coupled $CsPbBrI_2$ nanocrystals, using techniques such as polarization-resolved pump-probe spectroscopy and excitation-dependent PL, also demonstrated the presence of EHL at room temperature.[34] Notably, as shown in Figure 3e–f, this study also revealed ASE from the EHL, highlighting its large optical gain and constant density. Moreover, EHLs have been observed in the superlattice of $CsPbBr_3$[55] and thin films of $FaPbBr_3$,[82] although in these investigations, EHL was found to be stable only at low temperatures, as shown in Figure 3g.

The existence of EHL in MHPs hints at the possibility of a quasi-thermal and correlated state of photoexcited carriers and could offer valuable insights into optical phenomena such as ASE and SF. However, to fully understand the underlying mechanisms driving the formation and stability of EHL in

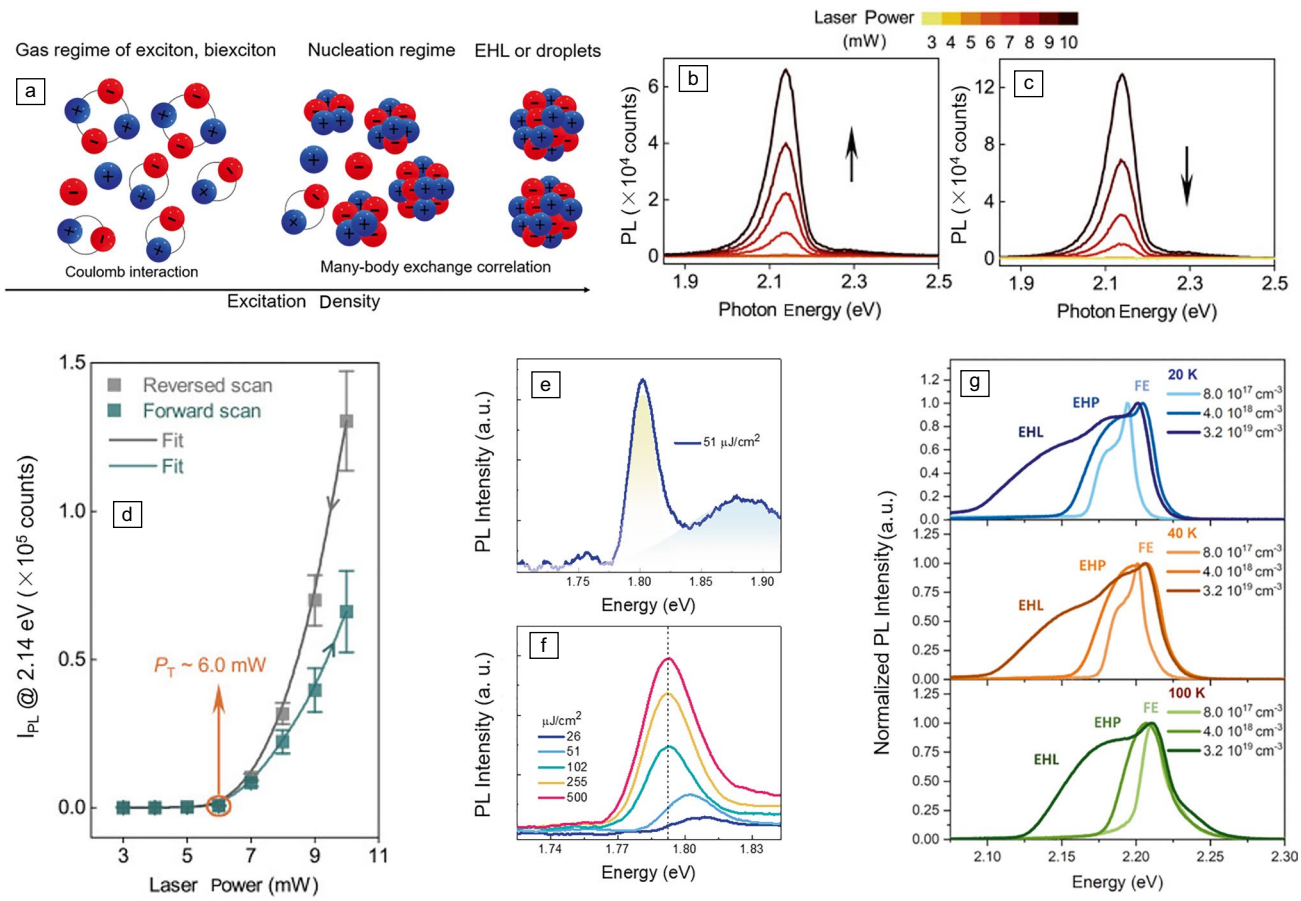

Figure 3. (a) Schematic illustration of electron–hole liquid (EHL) formation as a function of excitation density.[34] (b) Photoluminescence (PL) spectra of $CH_3NH_3PbBr_3$ single crystal as an increase in pump fluence.[81] (c) Decrease in pump fluence.[81] (d) Optical hysteresis curve demonstrates the signature of EHL formation.[81] (e) and (f) show the PL spectra of $CsPbBrI_2$ film, revealing amplified spontaneous emission (ASE), where, above 51 μJ/cm^2 a sudden shift takes place in the ASE position, and after that, position and linewidth remain invariant due to EHL formation.[34] (g) Excitation depended PL spectra of $FAPbBr_3$ thin film at different temperatures showing the EHL emission band.[82] FE, free exciton; EHP, electron–hole plasma.

MHPs, further studies are required. Additionally, a thorough investigation into the correlation between EHL formation and device performance parameters, such as efficiency and stability in optoelectronic devices, is essential for harnessing the full potential of EHL in MHP-based technologies.

Here, it is worth mentioning that SF and EHL are two exotic many-body phenomena of semiconductors that require protection from thermal decoherence, carrier–carrier scattering, and motional fluctuation. Observation of both these two states suggests that MHPs have some immunity against the source of decoherence, and the origin of such protection is indeed a topic of great interest and investigation. Some studies suggested that EPI can be one such mechanism by making a stable ground state and offering a shock absorber.[34,70] For instance, in the low carrier density regime, where polarons are stable entities, it could lead to the unusually long superradiant phase transition time, which is two orders larger compared to other semiconductors. Such coupling of electronic carriers paves the way for the macroscopic superradiant phase through vibration isolation. On the other hand, in the high carrier density regime, where although polarons and excitons are unstable entities, EPI can derive the system into a phase of quasi-thermal correlated electron–hole plasma that provides a suitable platform for superradiant phase transition.

Exciton–polariton condensation

Similar to other bosons, exciton–polaritons can undergo a remarkable phase transition of Bose–Einstein condensation (BEC). At sufficiently low temperatures or high carrier densities, a large fraction of these polaritons condense into the same quantum state, behaving as a single coherent entity.[51] However, unlike traditional BECs in atomic systems, exciton–polariton condensates exist in a nonequilibrium state due to the short lifetime of polaritons.[51]

Robust exciton–polariton with room-temperature stability makes MHPs attractive material for the realization of BEC. The polariton condensation in MHPs was first realized in 2017 using $CsPbCl_3$ microcavity structure (**Figure 4**a), where polariton lasing was demonstrated (Figure 4b–c) at room temperature.[83] Further, long-range (up to 60 μm) polariton condensation, manipulation, and formation of exciton–polariton condensate using electric fields were achieved using all inorganic MHP $CsPbBr_3$.[84,85] Mechanical exfoliation paves the way for the polariton condensation in 2D perovskites $[C_6H_5(CH_2)_2NH_3)_2PbI_4]$.[86] An important accomplishment using the MHPs as an active material is the realization of exciton–polariton condensation in the artificial potential landscape at room temperature, which was otherwise constrained to low temperatures.[50,87] By using the one-dimensional perovskite

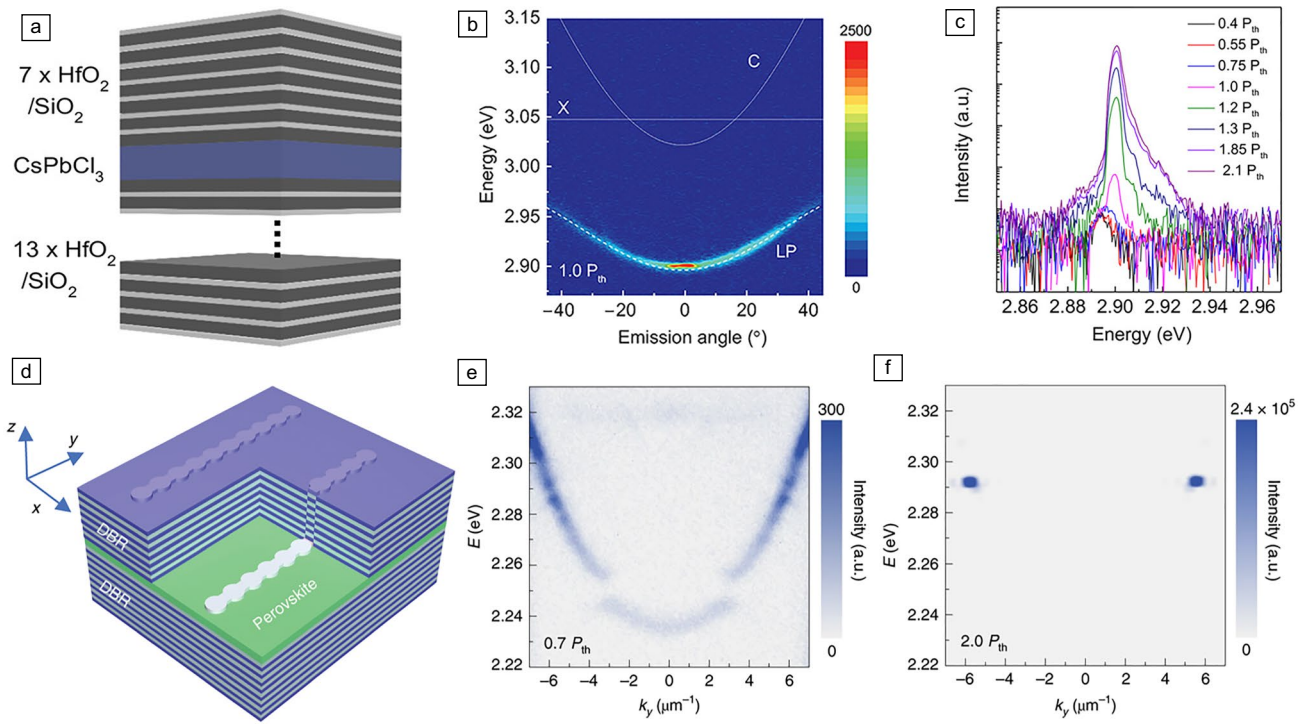

Figure 4. (a) Schematics showing microcavity structure of $CsPbCl_3$ crystalline nanoplatelet.[83] (b) Angle-resolved photoluminescence spectrum showing the onset of polariton lasing.[83] C, cavity; X, exciton; LP, lower polariton. (c) Ground-state emission spectra at $k\|=0$ show the transition to the nonlinear regime in the form of a sharp increase in intensity.[83] (d) Schematic diagram of 1D $CsPbBr_3$ lattice.[87] DBR, distributed Bragg reflector. (e) Polariton dispersion of 1D perovskite lattice.[87] (f) Polariton dispersion in the condensation regime demonstrates corresponding macroscopic occupation.[87]

lattice, as shown in Figure 4d, polariton condensation was achieved in p_y orbital states, as shown in Figure 4e–f.

Floquet states

The light-driven manipulation of electronic states is an exciting era of current research, where light fields are used to alter the absorption and emission of light. One prime example is light–matter hybridization, which arises from the time-periodic driving of electronic systems by the optical field. It offers an exciting avenue for coherent manipulation of the energy levels in semiconductors. As presented in **Figure 5**a, on nonresonant driving of a two-level system with energy gap E_0, no real population of the system is created, but coherent absorption and emission of light create the photon-dressing of states.[88] The interaction between electronic and Floquet states results in coherent modification of energy spacing by energy difference ΔE, which is known as OSE. The excitons with strong light–matter interaction, narrow bandwidth, and high oscillator strength in MHPs offer an exciting opportunity to study the Floquet states and coherently manipulate the excitons. In fact, in recent studies, MHPs have been found to be a strong candidate for exploring the Floquet states and their application in opto-spin and spintronics devices, even up to room temperature.[23,48,88–90] The spin-selective[88] (Figure 5b–c) and polarization dependence[89] of OSE in MHPs offers the ability to manipulate the spin states, making them valuable for applications in spintronics. In the layered 2D perovskite, OSE-induced energy shift and Rabi energy can be tuned by changing the dielectric contrast,[88] as shown in Figure 5d. For this purpose, the organic and halide components were substituted to change the dielectric constant of the barrier and well layers. In MHP quantum dots, the tuning of OSE is achieved by the facile method of size (Figure 5e) and composition tuning. Further, to tune the nature of shifting, biexciton states are explored in the quantum dots and 2D perovskites,[23,48] where blueshifted to splitting and redshifted OSE are demonstrated by changing the excitation energy. As shown in Figure 5f, by changing the driving energy, the strong exciton–biexciton interaction leads to the redshift to blueshift OSE.

Future of MHPs in quantum devices

The tunable collective and cooperative action of carriers in MHPs, including exciton–polarons, condensation, Floquet states, SF, and superradiance, makes them an ideal candidate for application in quantum technologies. These unique many-body states offer a pathway toward compact quantum

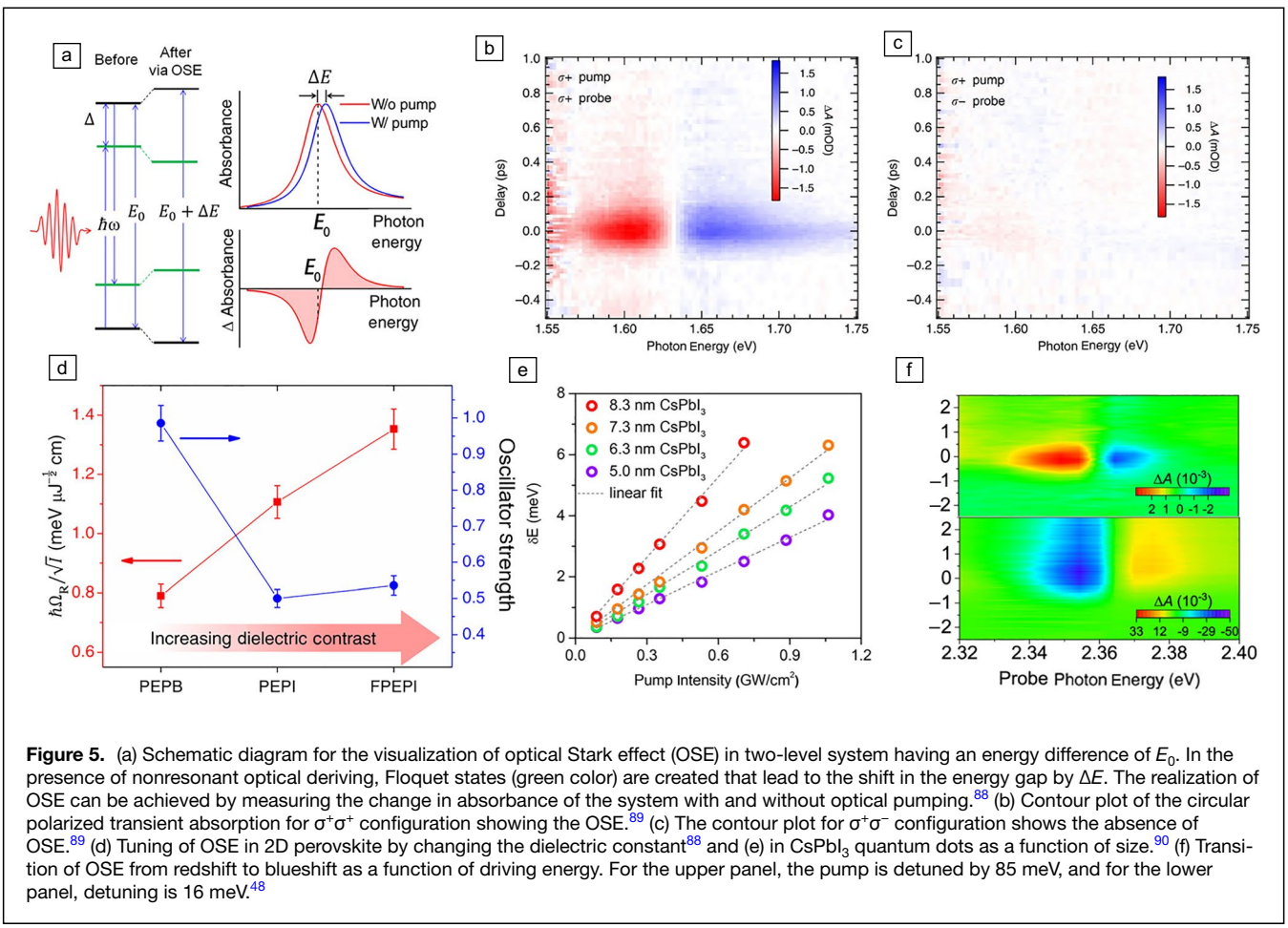

Figure 5. (a) Schematic diagram for the visualization of optical Stark effect (OSE) in two-level system having an energy difference of E_0. In the presence of nonresonant optical deriving, Floquet states (green color) are created that lead to the shift in the energy gap by ΔE. The realization of OSE can be achieved by measuring the change in absorbance of the system with and without optical pumping.[88] (b) Contour plot of the circular polarized transient absorption for $\sigma^+\sigma^+$ configuration showing the OSE.[89] (c) The contour plot for $\sigma^+\sigma^-$ configuration shows the absence of OSE.[89] (d) Tuning of OSE in 2D perovskite by changing the dielectric constant[88] and (e) in $CsPbI_3$ quantum dots as a function of size.[90] (f) Transition of OSE from redshift to blueshift as a function of driving energy. For the upper panel, the pump is detuned by 85 meV, and for the lower panel, detuning is 16 meV.[48]

light sources, ultrafast switching, novel quantum sensors, and components for quantum information processing by giving rise to the emergence of exotic phenomena that are not possible within a single particle frame.

The formation of hybrid exciton–polaron particles in MHPs offers the potential for enhanced interactions through polaron–polariton formation[91] and exploration of the OSE.[92] The attractive interactions between polarons could lead to advancements in sub-Poissonian light generation[57,58,92] for precision measurements, quantum communication, and imaging.

The realization of low threshold ASE, SF at room temperature, and single photon superradiance from individual quantum dots marks an exciting frontier for compact light sources. The ability to efficiently generate collective and cooperative emission with the ability of tunability across a wide spectrum creates the possibility of chip-based coherent quantum light sources, ultrashort pulse generation, superfluorescent lasers without resonant cavities, and remote sensing. The cooperative interactions or radiative coupling of emitters hold promise for reducing lasing thresholds and bypassing Auger recombination. Further, the formation of EHL offers a new mechanism for lasing and ASE due to the inherent constant density of carriers in EHL. It acts to keep the linewidth and position of emitted light invariant unlike ASE from the gas phase of exciton, biexciton, and electron–hole plasma where any change in carrier population affects the carrier density, hence properties of emitted light. Even though MHPs have emerged as active materials for achieving exciton–polariton lasing, ASE, SF, and superradiance, their current demonstration often relies on ultrafast (femtoseconds or picoseconds) optical excitation. There are very few studies that incorporate the CW and electrical excitation. Therefore, for widespread device integration, future research should focus on achieving these effects with either incoherent light or electrical excitation.[93]

An interesting regime of research in MHPs that is seeking attention is the self-assembly of quantum dots or nanocrystals.[67,68,94] Using MHP quantum dots as a building block, synthetic solid or multidimensional quantum dot arrays can be prepared with a plethora of applications, such as quantum simulators. Compared to naturally occurring quantum materials, which are often complex and challenging to control, quantum dot arrays offer an unprecedented degree of flexibility in engineering materials that exhibit exotic quantum phenomena such as Dirac cones, topological edge states, and nontrivial flat bands.[95,96] For instance, arranging quantum dots into a honeycomb lattice can mimic the electronic properties of graphene, leading to the emergence of Dirac cones. In these cones, the relationship between electron energy and momentum becomes linear, making electrons behave as if they are massless. This translates into ultrahigh electrical conductivity and novel electronic behavior, potentially suitable for high-speed, low-power devices or even new kinds of quantum computing. Further, the topology of the honeycomb array, coupled with spin–orbit interactions, can give rise to topological edge states.

Although the prospect of such an application is very compelling, the creation of perfectly ordered quantum dot arrays with the necessary precision is very challenging. In this regard, with a high degree of defect tolerance and size homogeneity, MHP quantum dots can turn the table and can offer a practical and real-life solution for the dream of an artificial solid with tunable properties. In fact, some efforts have already been made in this direction,[55] where superlattice of MHP quantum dots has been purposed as a candidate for the simulation of various quantum effects such as metal to insulator transition, random to cooperative emission by just changing the number of photoexcited carriers. In conclusion, we can say that along with their well-established application in photovoltaics and optoelectronic devices, MHPs are emerging as a strong candidate for solution processed quantum material.

Author contributions

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Conflict of interest

All the authors declare that they do not have competing interest.

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