RESEARCH HIGHLIGHTS

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The physical origin of stimulated emission in perovskites

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Perovskite lasers, due to their superiority in feasible production and wavelength tunability, find application in optical communication[1]. Since the discovery of stimulated emission from $CsPbCl_3$ microcrystalline at liquid-nitrogen temperature[2], successive breakthroughs in perovskite lasers have been made. In 2014, wavelength-tunable stimulated emission was realized by changing the anions of ABX_3[3], while random lasers in $MAPbI_3$ microcrystals were investigated for speckle-free imaging[4]. The threshold at that time was typically 10 $\mu J/cm^2$ under femtosecond laser excitation. In 2015, solution-processed nanowires significantly reduced the threshold to 220 nJ/cm^2[5]. With a high two-photon absorption, nanocrystals play a part in biological imaging and optical encryption[6]. $MAPbI_3$ films with distributed Bragg reflector (DBR) structures delivered a continuous-wave lasing with a threshold of 17 kW/cm^2[7]. In 2018, quasi-2D perovskites with quantum-well structure and improved stability attracted tremendous attention, which may perform as gain medium in amplified spontaneous emission[8] and lasing[9]. In 2020, $MAPbBr_3$ metasurface switched vortex lasing to linear polarized lasing within 1 ps, realizing terahertz ultrafast optical switches at low energy consumption[10]. Simultaneously, Adachi *et al.* applied triplet management strategies to overcome the lasing death phenomenon and realized room-temperature quasi-2D CW lasing[11]. Stimulated emission in perovskites is summarized in Fig. 1. Poor stability and unachievable mass production are two challenges for the commercialization of perovskite lasers[12].

Though much has been done in achieving low threshold and stability of perovskite stimulated emission, the mechanism remains controversial due to the coexistence of free carriers, excitons, exciton-polaritons, etc.[13]. Understanding the physical origin of stimulated emission in perovskites will inspire the manipulation on the lasing behavior for advanced applications. Exciton-polaritons, superfluorescence (SF), electron-hole plasma (EHP) and biexcitons may induce perovskite lasing.

In a strong-coupling regime, including well-constructed cavities like DBR and nanowires, photons are confined and strongly coupled to excitons[18] (Fig. 2(a)). The bosonic quasiparticles, known as exciton-polaritons, originate from the superposition of cavity photons and excitons, in which the weights of each faction are calculated to be Hopfield coefficients. The dispersion curves of polariton are anti-crossing (Fig. 2(b)). Because of the half-light half-matter property, the effective mass of polaritons is extremely small, typically 10^4 times smaller than that of a free electron[19]. Bose-Einstein condensation (BEC) of polaritons is supposed to take place even at room temperature[20], which brings pure quantum effect to a macroscopic scale. The massive occupation of the lower polariton branch relaxes in radiation within sub-nanosecond timescale, generating a large number of coherent photons (Fig. 2(c)). Since population inversion is not a necessity for polariton emitting, the threshold can be an order lower than ordinary lasers[21]. Following the observed polariton condensation in $CsPbCl_3$ nanoplatelets[14], investigations on polariton lasers towards low threshold and electrical pumped capability were reported in 3D[22], quasi-2D[9] and 1D[23] perovskites. Compared with GaAs, GaN and monolayer transition-metal dichalcogenides (TMDs), perovskites are the most suitable candidates to realize polariton devices with convenient fabrication and large binding energy[22]. Structures were designed to realize more sophisticated functions. In 2019, Rydberg exciton polaritons, with the excitonic constituent remaining in excited states, were first observed in a $CsPbBr_3$ cavity[24]. Polariton systems involving higher-order excited states built the foundation for exploring many-body physics in quantum optical information process. Liu *et al.* found that the condensation at various polariton states could be simply driven by changing the pump intensity in $CsPbBr_3$ microflakes, which provided the possibility of manipulating polariton states[25]. In 2021, a zigzag perovskite lattice, performing polarization-oriented optical switching, opened the gate of topological polariton devices[26]. Taking advantage of the delocalized and strongly interacting polaritons, on/off propagation states of the polariton fluid along a $CsPbBr_3$ microwire could be switched in picosecond timescale[27]. Developing polariton lasers provides a platform to investigate many-body physics, as well as to realize low-threshold polariton lasers and all-optical polariton logic devices[28]. Considering its great theoretical and practical significance, polaritons in perovskites will be a

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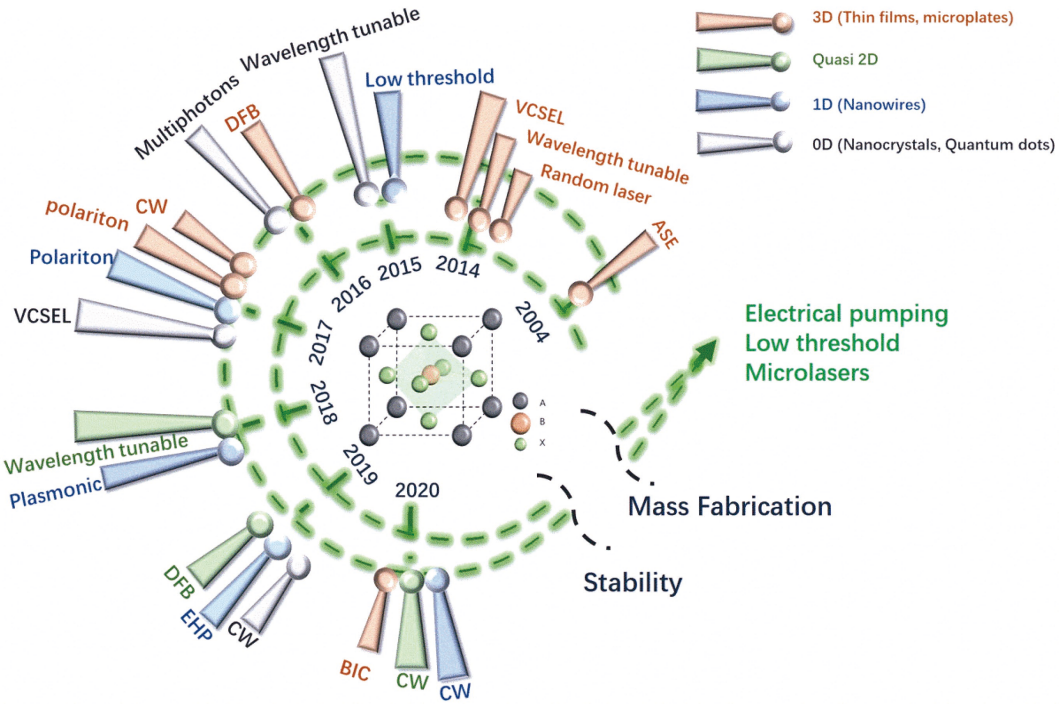

Fig. 1. (Color online) Advances of stimulated emission in 3D, quasi-2D, 1D and 0D perovskites.

hotspot.

Superfluorescence starts when the excited carriers spontaneously build up a macroscopic coherence under the material's polarization field[29]. Intensive bursts of light shows a strengthened first and second-order coherence, which enables the emission as entangled multi-photon quantum light source[15]. SF process requires a long dephasing time to obtain coherence among excited carriers. A collective emission from a fully incoherent system was more common, which is called amplified spontaneous emission (ASE)[30]. In SF dynamics, the time for the initially incoherent dipoles to form uniform orientation and build macro coherence is defined as delay time (τ_D) (Fig. 2(d)). In perovskites, most SFs were reported below liquid-nitrogen temperature, as $MAPbI_3$ thin films[31] and quantum-dot superlattice[15] were proved to perform SF emission. The delay time for SF is several picoseconds in both structures, and decreases at a higher excitation intensity (Fig. 2(e)). A strengthened first and second-order coherence identified that the emission originates from coherent excited emitters. Based on Rainò's work[15], a more detailed theoretical analysis on thermal decoherence and decay indicated a possibility to realize high-temperature SF[32]. SF may realize non-cavity, low-threshold and coherent high-quality light source.

Under intense excitation, the formation of excitons in perovskites is weakened by the screening effect, yielding plasma (Fig. 2(f)). Mott density was calculated to be 2×10^{17} cm^{-3} in $CsPbBr_3$[33]. A weakened dielectric response and bandgap renormalization induced by EHP have a significant impact on the lasing emission[34]. Time-resolved studies of $CsPbBr_3$ nanowires (Fig. 2(g)) revealed transient red shifting of lasing peaks and blue shifting of lasing gain profile, which could be attributed to the decay of EHP density[16]. The same phenomena of lasing peaks and gain profile were observed at a shorter time scale in two-photon pumped lasing dynamics[35]. Non-hybridized plasmonic lasing in $MAPbBr_3$ nanowires presented a rapid decay, which originated from EHP[36]. Recently, a quantitative analysis based on Drude-like model interpreted the transient shift, suggesting to suppress the density-induced shift by optimizing cavity geometry[37].

Biexcitons, originating from strong Coulomb interaction between two excitons, affect perovskite lasing properties. The many-body interaction among excitons usually causes a redshift in band-edge transition with a pump-intensity independency, typically quantified as a dozen meV[38]. The small binding energy makes biexcitons less resistant to thermal dissociation, which requires a low temperature for observation[34]. The radiative decay of biexciton presents an extra peak at the red side of free exciton in spectra (Fig. 2(h))[17]. The biexciton lasing in perovskites was first realized at 16 K with a threshold of 20 kW/cm^2[39]. By analyzing the dual-band emission below 160 K, excitons and biexcitons in $CsPbBr_3$ films performed a competitive mechanism[40]. Through many-body interaction, multiple excitons greatly affect the optical gain in perovskites. Xiao *et al.* applied 2D electronic spectroscopy and concluded that the optical gain in $CsPbBr_3$ nanocrystals was generated from strongly-interacting biexcitons[41]. Being treated with the mixture of $PbBr_2$, oleic acid and oleylamine, $CsPbBr_3$ nanoparticles realized a low-threshold trion gain with an increased stability, where trion is a charged exciton consisting of two electrons and one hole (or two holes and one electron)[42]. In Ruddlesden–Popper (RPP) perovskite, the biexciton lasing was realized in a vertical cavity with working temperature up to 125 K and a 50 meV binding energy[17]. Through transient absorption spectroscopy, Kanemitsu *et al.* claimed that the optical gain could be tuned by changing the excess energy of "hot biexcitons", which is composed of an exciton at the band edge and a hot exciton generated by pump pulse[43]. Since the presence of biexcitons and multiexcitons in perovskites are inevitable, especially in low-dimensional structures[17], more efforts should be made to clarify their influence on photon emission and Auger recombination process.

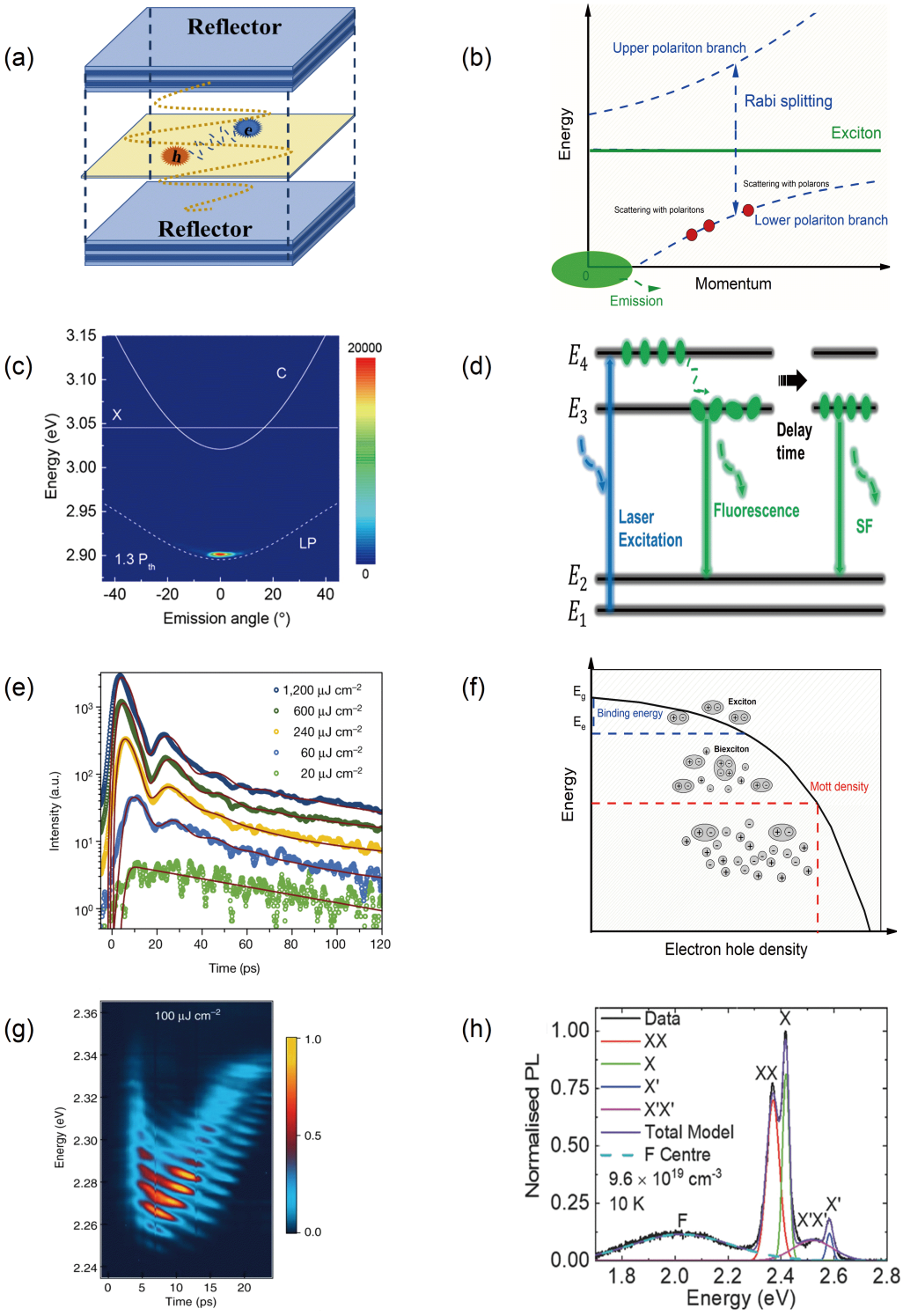

Fig. 2. (Color online) (a) The coupling between photon and exciton in the cavity. (b) The dispersion curves for polaritons. (c) Angle-resolved photoluminescence spectrum measured above the lasing threshold. The ground state is massively occupied, symbolizing polariton condensation. Reproduced with permission[14], Copyright 2017, American Chemical Society. (d) SF in a typical four-energy-level system. (e) Time-resolved SF emission of $CsPbBr_3$ QDs. Reproduced with permission[15], Copyright 2018, Springer Nature. (f) Exciton and EHP states with increasing electron-hole density. E_g and E_e refer to bandgap energy and exciton energy, respectively. (g) Time-resolved lasing of a single $CsPbBr_3$ nanowire at 80 K. Reproduced with permission[16], Copyright 2019, Springer Nature. (h) PL spectrum at 10 K (black trace) and Gaussian fits to various peaks. X and XX refer to the emission of exciton and biexciton, respectively. X′ and X′X′ refer to the emission of exciton and biexciton in other phases. Reproduced with permission[17], Copyright 2018, Wiley.

Ultrafast dynamics method can be used to study stimulated emission[16, 18]. Figuring out the physical origins of stimulated emission will help us to develop efficient and stable perovskite lasers.

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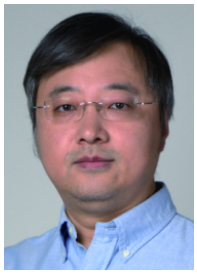

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