

Interaction of moiré excitons with cavity photons in two-dimensional semiconductor hetero-bilayers

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Abstract: Moiré materials, composed of two single-layer two-dimensional semiconductors, are important because they are good platforms for studying strongly correlated physics. Among them, moiré materials based on transition metal dichalcogenides (TMDs) have been intensively studied. The hetero-bilayer can support moiré interlayer excitons if there is a small twist angle or small lattice constant difference between the TMDs in the hetero-bilayer and form a type-II band alignment. The coupling of moiré interlayer excitons to cavity modes can induce exotic phenomena. Here, we review recent advances in the coupling of moiré interlayer excitons to cavities, and comment on the current difficulties and possible future research directions in this field.

Key words: moiré interlayer excitons; optical cavity; exciton–polariton; Bose-Einstein condensation

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1. Introduction

Since the discovery of graphene, the field of two-dimensional (2D) materials has become one of the most exciting and fastest-growing fields in condensed matter physics. Many exotic phenomena have been discovered in 2D materials and the van der Waals (vdWs) heterostructures composed of them^[1–4]. This is enabled by reliable dry transfer techniques that construct multilayer structures while maintaining clean interfaces between layers^[5]. When two vdWs materials are stacked vertically, a moiré material is formed if the two materials have a lattice mismatch or are stacked with a finite twist angle. In this case, in the 2D plane, the atoms are arranged to form periodic patterns called “moiré patterns”, whose lattice constants are much larger than the intrinsic lattice constants of the two layers (Fig. 1(a)). The slowly changing local atomic configuration in space causes the energies of particles such as electrons and excitons to change slowly, forming a moiré potential in the material^[6–8] (Fig. 1(b)). Particle motions in this potential have relatively low energies and are therefore distinguished from “high-energy” physics characterized by the scale of the intrinsic lattice constant of individual layers. Another effect of the moiré pattern is the folding of the Brillouin zone, forming moiré bands, which is helpful for the study of strongly correlated physics in moiré systems^[9, 10].

An important material to construct moiré materials is the transition metal dichalcogenide (TMD) semiconductors. Single-layer TMD semiconductors have direct bandgaps at the K and K' points, making them suitable materials with

bright excitons^[11]. The lattice constants of the TMDs are similar, with the largest lattice mismatch being around 13%, which makes the TMD family a good material for constructing moiré materials. By controlling the twist angle, a wide range of moiré lattice constants can be achieved. The moiré hetero-bilayer formed by TMDs can also support normal intralayer excitons. However, if we carefully choose TMD materials to form a type-II band alignment (Fig. 1(c)), the hetero-bilayer can support a new type of excitons called interlayer excitons^[12]. The electrons and holes in the interlayer excitons are located in different layers, respectively, and thus, the interlayer excitons can strongly sense the moiré potential. Furthermore, the lifetime of interlayer excitons is much longer than that of intralayer excitons due to the small overlap of electron and hole wave functions^[12]. The separation of electrons and holes provides permanent out-of-plane dipoles of interlayer excitons, which enables Stark tuning of exciton energies^[13] and enlarges the interactions between excitons^[14]. These properties make the TMD-based moiré systems good platforms to study strongly correlated physics of excitons.

When excitons are placed into a near-resonance cavity, they naturally couple with cavity modes. At weak coupling regime, we can observe the Purcell effect, i.e., enhanced photon emission and shortened exciton lifetime. A new type of quasiparticle called an exciton–polariton emerges when the system enters strong coupling regime, where the coupling strength between the excitons and photons is greater than the linewidth. The most common systems for studying exciton–polaritons are semiconductor quantum wells, and TMDs have also been extensively studied in recent years. Many outstanding discoveries and applications have been made in exciton–polariton systems, such as Bose-Einstein condensation (BEC)^[15], superfluidity^[16], and low-threshold exciton–polariton lasers^[17]. Exciton–polariton systems have become

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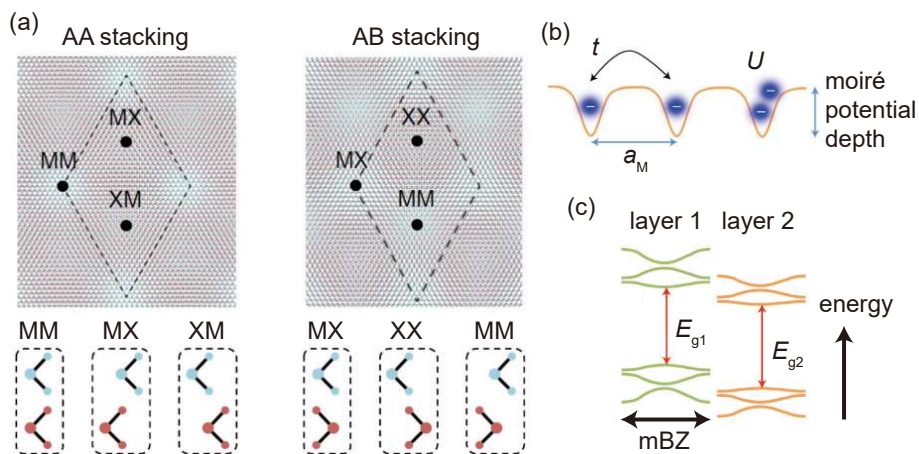


Fig. 1. (Color online) Moiré lattice. (a) Moiré lattice structure for AA-stacked and AB-stacked TMD semiconductors (top). The high-symmetry sites for each case are labelled; their cross-section views are shown (bottom). The large and small dots label the transition metal atom (where M is Mo and W) and the chalcogen atom (where X is S, Se and Te), respectively. (b) Schematic illustration of an array of moiré atoms that trap electrons, which can tunnel between neighboring sites with amplitude t and experience on-site Coulomb repulsion U . (c) Schematic layer-resolved moiré band structure for semiconductor moiré materials with type-II band alignment. mBZ stands for mini-Brillouin zone, and E_{g1} and E_{g2} are the bandgap of the first and second TMD layers, respectively^[6].

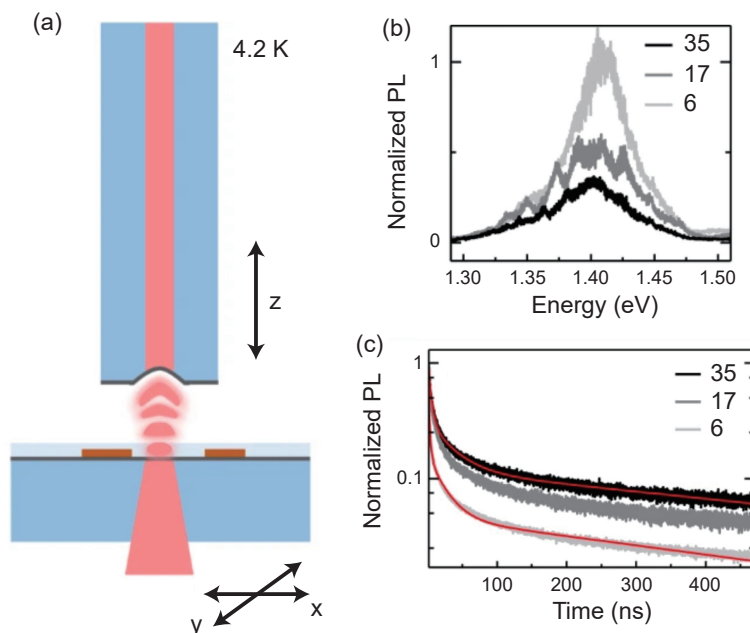


Fig. 2. (Color online) Moiré exciton in a FP cavity. (a) Cavity setup at 4.2 K: the fiber-based micro-mirror forms the cavity together with a planar macro-mirror with CVD-grown MoSe_2 - WSe_2 heterostructure on top. Independent translational degrees of freedom enable lateral sample displacement and cavity length detuning. (b) Spectra of interlayer exciton PL for the corresponding cavity lengths. (c) Traces of interlayer exciton PL decay shown for three selected cavity lengths of 35, 17, and 6 μm . The solid lines are fits to the data with three-exponential decay constants. Note the speed up in the decay upon the reduction of the cavity length^[19].

one of the main tools for studying and manipulating light-matter interactions. As mentioned above, due to the long exciton lifetime, the out-of-plane dipole, and the strong interaction with the moiré potential, we expect to discover new exotic phenomena and better control when the moiré excitons are strongly coupled to the microcavities. Of course, like normal excitons, moiré excitons can also be coupled to Fabry-Pérot (FP) cavities or photonic crystals.

2. Moiré excitons in FP cavities

A typical system supporting moiré excitons is a heterostructure composed of two single-layer TMD semiconductors, which is naturally flat and can be integrated into a vertical FP

cavity. When a hetero-bilayer is embedded into an FP cavity, the light-matter coupling within the cavity leads to the formation of exciton-polaritons, and strongly modify the spectrum of the hetero-bilayer^[18]. These hetero-bilayers were first integrated into an FP cavity in 2019^[19], where a CVD-grown AB stacked MoSe_2 - WSe_2 hetero-bilayer was placed into the FP cavity. The FP cavity has a fixed flat mirror made of Si/SiO₂ substrate and a sphere fiber-based movable top mirror, so the lateral sample position and cavity length can be controlled separately (Fig. 2(a)). The existence of interlayer excitons in the bare hetero-bilayer is confirmed by its photoluminescence (PL) and differential reflectance spectra. When placed into the cavity, the MoSe_2 - WSe_2 sample couples with the cav-

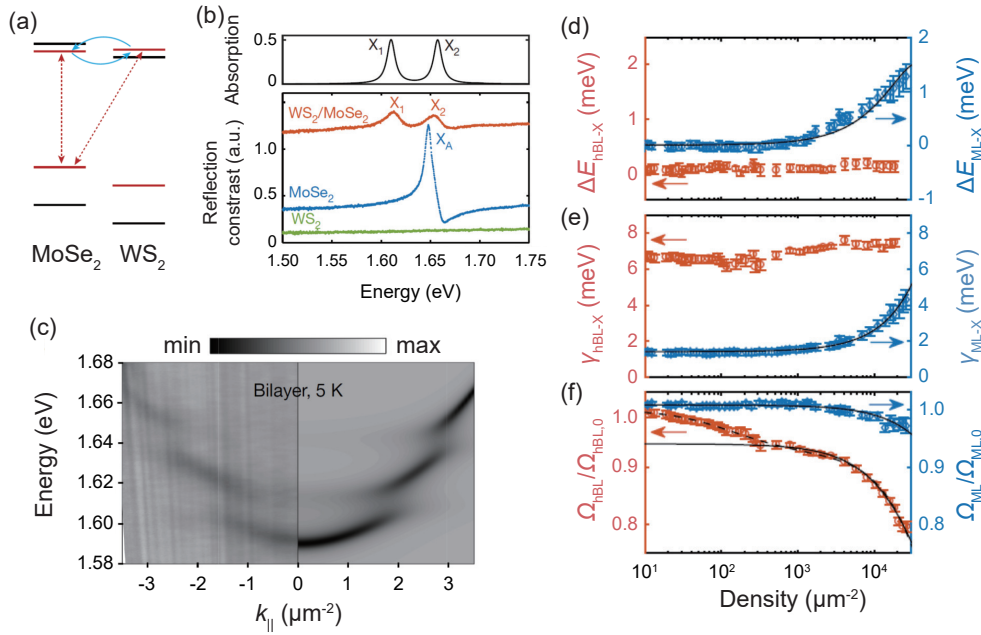


Fig. 3. (Color online) Hybrid moiré exciton in a FP cavity. (a) Resonantly enhanced interlayer hybridization of TMD double layers can result from interlayer charge hopping (blue arrows) in conduction bands for certain combinations of TMDs. Such band alignment yields nearly degenerate intra- and interlayer exciton species (red arrows)^[21]. (b) Top, theoretical optical absorption spectrum calculated from the moiré exciton band structure. Bottom, reflection contrast spectra near the MoSe₂ A-exciton resonance, from the WS₂–MoSe₂ hetero-bilayer (red), MoSe₂ monolayer and WS₂ monolayer (green). The MoSe₂ monolayer A-exciton (X_A) splits into two well resolved moiré excitons (X_1 and X_2) in the hetero-bilayer. (c) Angle-resolved white-light reflection spectra, demonstrating strong coupling between moiré excitons and cavity photon at 5 K. The left and right panels show the measured and simulated results, respectively. (d–f) Shift of exciton energy ΔE , half-linewidth γ , and normalized coupling strength Ω/Ω_0 of the hetero-bilayer LPs (red) and monolayer LPs (blue) as a function of carrier density. The hetero-bilayer $\Delta E_{\text{HBL-X}}$ and $\gamma_{\text{HBL-X}}$ (red circles in (d, e)) are approximately constant. The monolayer $\Delta E_{\text{ML-X}}$ and $\gamma_{\text{M-X}}$ (blue diamonds in (d, e)) are fitted by a second-order polynomial and a linear fit, respectively (black solid lines in (d, e)). In (f), the black lines and black dashed line are fitting results^[20].

ity mode, resulting in a Purcell enhancement of its PL. As shown in Fig. 2(b), as the cavity length is shortened from 35 to 6 μm , the PL intensity of the sample increases exponentially. Fig. 2(c) shows the time-resolved normalized PL at different cavity lengths. It can be found that the decay rate increases with the decreasing cavity length. These phenomena suggest the Purcell enhancement of exciton emission induced by the microcavity. The light-matter coupling strength of interlayer excitons can be extracted from the time-resolved PL data and found to be two to three orders of magnitude smaller than that of single-layer TMDs, which is consistent with the nature of spatially separated electrons and holes in interlayer excitons. The low light-matter coupling strength prevents the strong coupling between interlayer excitons and cavity modes, hindering the further development and application of such systems.

To overcome this intrinsic difficulty, Zhang *et al.* uses hybrid moiré excitons instead of pure interlayer/moiré excitons to couple to cavity modes^[20]. When the materials in the hetero-bilayer have nearly identical conduction band minima (or valence band maxima), hybrid moiré excitons are formed, i.e., the electrons (or holes) of the excitons can hop between the two layers, forming a mixed state consisting of intralayer and interlayer excitons^[21], as shown in Fig. 3(a). In this work, they placed a WS₂–MoSe₂ hetero-bilayer with a twist angle of 56.5° at the antinode of an FP microcavity. The hetero-bilayer can support two kinds of hybrid moiré excitons with energies around 1.61 and 1.65 eV, respectively (Fig. 3(b)). These hybrid moiré excitons strongly couple with

cavity modes and form exciton–polaritons. The angle-resolved white light reflectance spectrum of this cavity clearly shows the exciton–polariton anti-crossing behavior, indicating the strong coupling of the excitons to the cavity modes (Fig. 3(c)). Then, by fitting the measured exciton–polariton dispersion using a three-coupled oscillator model, the coupling strength between the excitons and the cavity is obtained. Based on the robust existence of moiré exciton–polariton, the authors investigated the influence of excitation density on the properties of exciton–polariton. Compared with normal 2D exciton–polaritons, the moiré exciton–polaritons show several distinct features. Generally, both the energy and linewidth of an exciton–polariton mode increase with the excitation density due to exchange interactions, dephasing, and Pauli blocking of excitons. However, in the case of moiré exciton–polaritons, the lower (LP) and middle (MP) exciton–polariton branches move symmetrically towards the lower moiré exciton energy (1.61 eV), while their linewidths do not change. Furthermore, fitting the measured dispersion using the coupled oscillator model can reveal the nature of excitons and uncover more differences between normal and moiré excitons. In normal systems, when the excitation density increases, the exciton energy, and linewidth increase, while exciton-photon coupling strength decreases. In sharp contrast, the extracted moiré exciton energies and linewidths are essentially independent of the excitation density, and the coupling strength is significantly lower than in the single-layer MoSe₂ case (Figs. 3(d)–3(f)). These phenomena are similar to those of quantum dot excitons, suggesting the zero-dimen-

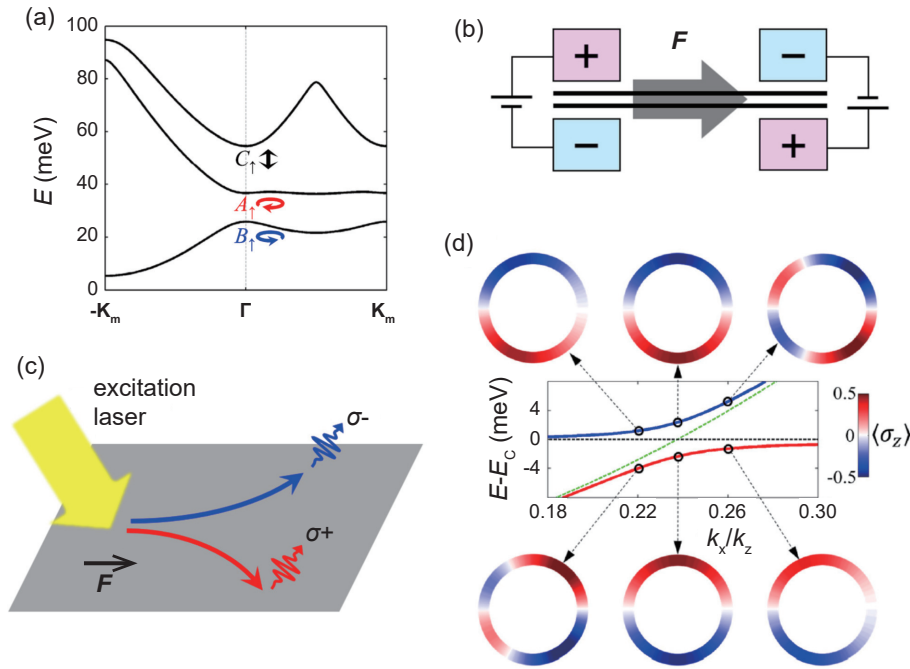


Fig. 4. (Color online) Novel theory prediction of moiré exciton in a cavity. (a) The three lowest bands of the spin-up moiré interlayer exciton with a 5 nm moiré period. The moiré potential leads to three well-separated eigenmodes B_{\uparrow} , A_{\uparrow} and C_{\uparrow} at Γ , which have σ^- , σ^+ and z -polarized optical dipoles, respectively. (b) The gradient of an interlayer bias introduced by a split gate can apply an in-plane force F on the interlayer exciton. (c) Illustration of the polarization Hall effect driven by F . (d) Scattering induced polarization currents under excitation at given energy-momentum points (empty dots) in the LP and UP branches. Cavity photon lifetime is taken as 1 ps^[22].

sionality of moiré excitons. In a moiré lattice, the wave function of the excitons localizes at the potential minimum. If two excitons are located at the same potential minimum, the interaction between them is very strong and larger than the exciton linewidth, i.e., exciton blockade occurs. Therefore, excitons tend to occupy potential wells alone, which makes the interactions between excitons small, as the amplitude decreases rapidly with distance. The absence of intra-site interactions results in the absence of energy shift or exciton-induced dephasing. At the same time, the abnormal decrease of coupling strength is also caused by the confinement of excitons. When all the moiré lattice is occupied by excitons, the exciton-photon coupling saturates, and the calculated critical density is much smaller than that in the single-layer case and leads to a rapid drop at high excitation densities. Additionally, at very low excitation densities, large nonlinearities and strong exciton saturation were found, suggesting some hidden mechanisms.

In addition to these experimental works mentioned above, theorists have also proposed some new properties of moiré interlayer excitons in a cavity, such as topological transport^[22]. In a moiré superlattice, the moiré potential leads to three well-separated interlayer excitons eigenmodes with left-handed in-plane (A_{\uparrow} mode), right-handed in-plane (B_{\uparrow} mode), and out-of-plane (C_{\uparrow} mode) optical dipoles, respectively (Fig. 4(a)). These resonances can selectively couple with differently polarized cavity modes. By carefully tuning the vertical position of the hetero-bilayer in the microcavity, the cavity modes can be decoupled from the first two (A_{\uparrow} and B_{\uparrow} modes) or the last (C_{\uparrow} mode) resonance, yielding C_{\uparrow} -polaritons and $A_{\uparrow}B_{\uparrow}$ -polaritons respectively. In the first case, the C_{\uparrow} -polariton has a k -dependent circular polarization (Fig. 4(d)). If the polaritons are driven by the in-plane force introduced by

the split gate (Fig. 4(b)), a polarization current is formed. In the other case, for the same k , the $A_{\uparrow}B_{\uparrow}$ -polaritons of different spins have opposite Berry curvatures. As a result, if the polaritons are driven by in-plane force, a polarized Hall current will appear (Fig. 4(c)). Interlayer moiré exciton-polaritons also exhibit optical nonlinearity if coupled to intralayer excitons^[23]. After considering the intra-site repulsion of excitons and the saturation effect of light-matter coupling, the authors found distinct characteristics compared with ordinary 2D polariton gases. When the system is coherently driven, the density optical bistability exists within a certain driving field, cavity-exciton detuning, and inter-site interaction energy range. The system also allows incoherent pumping, such as optically driven higher exciton bands that rapidly relax to the hybrid state. With a maximum occupation number of 2 per site, it is found that the system allows either one-photon or two-photon processes, which means that the system has the potential to realize single- and multi-photon lasers. All of the above features arise from strong intra-site interactions that cannot be predicted by the Gross-Pitaevskii equation commonly used in the study of normal 2D polariton gases.

Theorists have not only predicted the properties of interlayer exciton-polaritons, but also studied intralayer exciton-polaritons in moiré systems, such as AA-stacked MoSe₂-WSe₂ hetero-bilayers^[24]. Although the intralayer excitons are confined within one layer, it can still sense the moiré potential and be confined to the potential minimum. The moiré potential also produces many exciton resonances in the mini-Brillouin zone. The exciton energy, localization, and coupling strength are simultaneously controlled by the twist angle. Therefore, by changing the twist angle and the cavity length, one can change the polariton energy and the number of polariton branches.

3. Moiré exciton in photonic crystals

The flexibility and atomically flat surface of the TMD hetero-bilayers also allows them to be integrated with other types of substrates, such as photonic crystals. By transferring the hetero-bilayer directly onto the cavity made of photonic crystals, the interlayer excitons can be effectively coupled to the cavity modes. Specially, Rivera *et al.* integrated a gallium phosphide (GaP) linear three-hole defect (L3) photonic crystal cavity with carefully aligned MoSe₂-WSe₂ hetero-bilayer with a twist angle of less than 5°^[25]. Compared with the uncoupled hetero-bilayer, the sample located in the cavity has two narrow and sharp peaks in its PL spectrum, corresponding to the resonances of the cavity. These narrow peaks are also strongly linearly polarized, corresponding to the linear polarization of the cavity modes. After accounting for the reduction in collection efficiency caused by the redistribution of the emission momentum vectors, the radiation rate enhancement is estimated to be about 60 times. In a similar structure, Paik *et al.* placed a WSe₂-MoSe₂ hetero-bilayer on a SiN grating cavity^[26]. The grating cavity has a large area and can completely cover the sample, allowing the sample to be fully coupled to the optical mode. In this way, they successfully realized lasing gained by moiré interlayer excitons and confirmed the large coherence length of the excitons. Similar to the case of FP cavities, although people have tried to integrate moiré interlayer exciton with photonic crystals, no strong coupling has been achieved so far. This may also originate from the low oscillation strength of the moiré interlayer excitons.

4. Summary and outlook

Compared with normal excitons, moiré excitons, especially interlayer or hybrid moiré excitons, have additional intriguing properties such as permanent out-of-plane dipoles, electrically tunable energies, long-range and strong interactions, and tunable spatial confinement by moiré potential. These properties make the moiré excitons an ideal platform to study many-body quantum phenomena. Moiré excitons alone have been intensively investigated, however, studies on their coupling to light are still limited. Most of the works mentioned above are at the weak coupling regime, failing to achieve strong coupling between moiré excitons and cavity modes. This perhaps is caused by the intrinsic low oscillation strength of moiré interlayer excitons, which is about two orders of magnitude lower than that of single-layer TMDs. As far as we know, so far, Zhang *et al.*'s work is the only one that has successfully entered the strong coupling regime^[20], in which hybrid moiré excitons are used instead of pure interlayer excitons to avoid this problem. Although many novel moiré exciton-polariton phenomena have been discovered, such as 0D behavior and large nonlinearities, the field still needs a lot of development.

One of the most important phenomena of exciton-polariton is BEC, which is the basis for many applications and phenomena of exciton-polariton, such as low-threshold laser and superfluidity. However, BEC in moiré exciton-polariton has not yet been realized. In the work of Zhang *et al.*, the coupling strength of moiré exciton rapidly decreases with increasing excitation density, preventing them from reaching the

BEC. This is caused by the confinement of the moiré potential and the strong interaction between excitons, which lower the critical density of saturation and leads to a rapid loss of coupling strength beyond this critical density. A possible solution is to increase the twist angle to lower the lattice constant of the moiré pattern, increase the hopping between moiré cells and reduce the confinement of excitons. However, the existence of bright hybrid moiré exciton requires tight control of the interlayer alignment to form a direct bandgap. In other words, the twist angle cannot be changed at will. Only a few twist angles suffice and make *k*-valleys of different layers overlap each other greatly^[24]. Another strategy is to use new materials with conduction band minima and valence band maxima at the center of the Brillouin zone, such as few-layer InSe^[27], which maintain a direct bandgap regardless of the twist angle. In this way, we can increase the saturation density and may achieve moiré interlayer exciton BEC.

The permanent out-of-plane dipoles of moiré interlayer excitons enable electric control of their energy by simply applying a gate voltage to them. Furthermore, if a spatially varying gate voltage is applied, we can control the flow of interlayer excitons and can fabricate exciton switches^[28]. In principle, the moiré interlayer exciton-polariton can also be controlled in this way because of the finite exciton part in it. Exciton-polaritons have been a good platform for quantum simulations^[29–31], but ideal simulators require easy and programmable control of the potential landscapes. The best way to construct such a potential now is by photoexcitation on the sample to form a spatially dependent exciton reservoir, then take the repulsion between exciton-polariton and exciton reservoir as the potential^[32]. However, the interactions between excitons and exciton-polaritons are intricate, and excitons in the reservoir can be converted into exciton-polaritons, bringing more complexity to the simulated system. Here, the moiré interlayer exciton-polaritons provide a new route to generate a programmable potential by adjusting the voltage of distributed gates. The electrostatic potential is relatively clean and does not produce complex interactions, which helps better simulate the target system.

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