Superradiant Emission from Coherent Excitons in van Der Waals Heterostructures

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Recent advancements in isolation and stacking of layered van der Waals materials have created an unprecedented paradigm for demonstrating varieties of 2D quantum materials. Rationally designed van der Waals heterostructures composed of monolayer transition-metal dichalcogenides (TMDs) and few-layer hBN show several unique optoelectronic features driven by correlations. However, entangled superradiant excitonic species in such systems have not been observed before. In this report, it is demonstrated that strong suppression of phonon population at low temperature results in a formation of a coherent exciton-dipoles ensemble in the heterostructure, and the collective oscillation of those dipoles stimulates a robust phase synchronized ultra-narrow band superradiant emission even at extremely low pumping intensity. Such emitters are in high demand for a multitude of applications, including fundamental research on many-body correlations and other state-of-the-art technologies. This timely demonstration paves the way for further exploration of ultralow-threshold quantum-emitting devices with unmatched design freedom and spectral tunability.

1. Introduction

Superradiant emitters (SREs) are a new generation of light sources in which an ensemble of identical emitters produces a standing-wave of electric-dipoles that stimulate the emission from the system via collective light-matter interactions under a common pumping source.\[1-4\] The synchronized dipole oscillation leads to a strong amplification of the system's emission\[5,6\] that maintains a robust phase coherence and spectral purity,\[7,8\] which are required to circumvent the existing challenges of spectral broadening in conventional lasers\[1,9\] and appealing for countless applications, including fundamental sciences and cutting-edge technologies.\[10-12\] SRE using an ensemble of suspended quantum dots, alkaline-earth atoms etc., has been achieved recently.\[2,4,13\] It is believed that obtaining SRE based on solid-state emitters can dramatically improve the state-of-the-art solid-state lighting devices' spectral purity, potentially circumventing the rigid requirement of phase coherence to address the ever-increasing demand for coherent light sources. However, the realisation of SRE using a semiconducting solid remains a great challenge.

Monolayer transition metal dichalcogenides (1L-TMDs) exhibit strong electromagnetic interaction and accommodate excitons of extraordinarily high binding energy even at room temperature.\[14-18\] Additionally, the quantum-well-like confinement produces 2D confined in-plane excitons under a photon field of the material's bandgap energy.\[19-21\] Recently, Stevens et al. proposed the theoretical prediction of superradiant coupling of excitons in monolithic TMD layers,\[22\] and experimentally demonstrated a superradiant absorption in bulk TMDs. However, to obtain superradiant emission, a direct bandgap is necessary. Although
1L-TMDs possess a direct bandgap, it suffers from huge surface damping, which significantly influences the emission intensity making it challenging to obtain superradiant coupling of the excitons. In our design, employing the dielectric spacer in TMD/hBN/TMD heterostructure, the hBN layer preserves the direct bandgap nature of TMD layers and reduces the surface damping of the excitons while keeping them in close proximity. With these unique advantages, we attempt a completely different approach to obtain ultra-narrow band SRE from a novel heterostructure composed of 1L-TMD and a few-layered hBN (FL-hBN). The collective exciton-exciton interaction via a common photon field in such system at low temperature produces coherent ultra-narrow band emission. Along with the generation of an ultralow threshold sharp spectrum utilising a simple design, our study opens a new avenue for the realisation of entangled quantum emitting devices with great freedom of structural scheme and emission energy tunability.

2. Results and Discussion

2.1. Device Design

Single crystalline 1L WSe₂ and FL-hBN were mechanically exfoliated on thin transparent polydimethylsiloxane (PDMS) membrane using a standard exfoliation technique and then transferred on SiO₂/Si substrates to design WSe₂/hBN/WSe₂ heterostructure (Figure 1A). Optical microscopy (OM) image of the designed WSe₂/hBN/WSe₂ heterostructure is presented in Figure 1B. The thickness of the TMD and hBN layers was confirmed by atomic force microscopy (AFM) imaging as shown in Figure 1C. The bright-, dark-field and fluorescent images of the heterostructure are provided in Figure S1 in the Supporting Information. A series of samples with different hBN thicknesses between WSe₂ layers was designed, and their performance was studied. The OM and AFM images of those samples are provided in Figure S2 in the Supporting Information. The spatial mapping of integrated photoluminescence (PL) intensity of the device in Figure 1B is shown in Figure 1D. The corresponding Raman spectroscopy mapping of the heterostructure is shown in Figure S3 in the Supporting Information, which further demonstrates the composition of the heterostructure.

2.2. Interlayer Exciton Coupling at 300 K

We first analyse the room temperature (300 K) PL spatial map over the sample, as shown in Figure 1D. The corresponding average PL signal over $2 \times 2 \mu m^2$ at the monolayers and across the heterostructure consisting of varying width of hBN spacers are shown in Figure 2A. Interestingly, we observed that the PL intensity is strongly amplified at the heterojunction depending...
on the hBN spacer thickness. The enhancement factor, defined as the ratio of integrated PL intensity at the heterostructure and the combined integrated intensity at the monolayers, \((I_H)/(I_{L1}+I_{L2})\), is shown as a function of hBN spacer thickness and measurement temperature (Figure 2B). These intriguing results can be understood in terms of the coupling and aggregation of interlayer excitons across the hBN layer,[23–26] where hBN acts as a spacer in between the excitons located at the individual monolayers (see Figure 2C). Exciton interaction becomes stronger as the separator thickness is reduced to only a few layers of hBN. It is worth mentioning that the coupling strength is significantly reduced when the hBN thickness increases beyond 10 nm. Indeed, hBN as a substrate plays a significant role in PL enhancement of the monolayer TMDs, which is mainly due to the protection of the hBN layer that strongly suppresses the physio- and chemisorption of impurities, providing an atomically flat surface for sample exfoliation to avoid ripples and preventing doping from the SiO2.[27,28]

2.3. Robust Exciton Coupling at Low Temperature

To further investigate the nature of exciton coupling, we have recorded the temperature dependence of PL emission spectra by cooling the sample down to the liquid helium temperature. Temperature-dependent mapping of the integrated PL emission intensity is shown in Note S3 in the Supporting Information. Suppression of electron-phonon interactions at a lower temperature enhances the coupling and, as a result, the PL intensity at the heterostructure is strongly enhanced. The corresponding plot of excitonic coupling strength (enhancement factor) at the heterostructure as a function of temperature is shown in Figure 2B. The emission from the heterostructure becomes more appealing at a temperature \(<20\text{ K}\), where multiple pronounced sharp peaks superimpose with the broad emission profile (Figure 3A). The spatial distribution of such sharp peaks over the heterostructure has been mapped, as shown in Figure 3B. Prior to the publication of these results, this intriguing phenomenon has not been observed in any TMD-based heterostructures. Intuitively, it can be understood as follows, and a more detailed discussion is provided in the “underlying mechanism” section. The excitons in the atomically thin 2D WSe2 at low temperature turn into an ensemble of optically pumped identical dipoles confined in the 2D quantum-well.[23,26,27,32] The reduction of the electron-phonon coupling at low temperature gives rise to the strong Columbic attraction among the excitonic dipoles, which promotes the stabilization of dipoles with the pumped photonic field.[16,26,33–35]
Such dipoles in two parallel 2D quantum-wells separated by a few-atom-thin insulating spacer induce a robust dipole-dipole correlation due to the collective light-matter interaction at the active WSe$_2$ layers in the heterostructure (see Figure 2C). As a result, a standing wave of the electric field due to the dipoles is generated. This stimulates a collective emission with an exceedingly high phase coherence.\[1,2,5–7,9,13,32,36,37\] Thus, we have observed the appearance of pronounced sharp peaks in the emission spectra and uniform distribution of these peaks over the sample area (except the part distorted by wrinkling and impurities) is obtained (Figure 3B,C,D). The enhanced phonon-exciton coupling at higher temperatures counteracts such dipole-dipole coupling, annihilating the synchronization of the oscillation and the sharp peaks vanish. Our finding supports the presence of interlayer dipole coupling and the uniform emission over the flake contrasts the previously reported point defects or localized exciton associated with single-photon emitters from monolayer WSe$_2$\[38–41\] in Notes S4 and S5 in the Supporting Information, we have provided a more detailed discussion on the obtained difference between the observed emission with single-photon emission and formation of Moiré excitons in WSe$_2$ based heterostructures, respectively.

2.4. Superradiant Emission

To unveil the nature of the sharp emission lines, we have recorded the emission spectra under different excitation power densities, as shown in Figure 4A. Interestingly, we observe the appearance of sharp peaks at a very small detectable pumping power density with a critical threshold of $\approx 0.8$ μW μm$^{-2}$. The integrated emission intensity of the peaks also increases super-linearly with the pumping power density, as shown in Figure 4B. The lower excitation power density produces a lesser number of excitons, and as the excitation pumping power density is increased, a larger population density of excitonic dipoles is generated, resulting in an abrupt enhancement of dipole-dipole coupling in the ensemble.\[1,6,8,13\] The collective oscillation of such photo-excited dipoles provides a large number of emitted photons with an exceedingly high phase coherence.\[1,2,6,9,36,37\] As shown in Figure 4B, the result shows...
a superlinear enhancement of the integrated intensity of the emitted photons and a superlinear reduction of the full width at half-maxima (FWHM) of the emission peak with pumping power density. The obtained linewidth of 0.1 nm (≈200 μeV) corresponds to a coherence length of ≈0.56 cm for the emitted lines from the device, comparable to the coherence length of a standard semiconductor laser with a well-designed optical cavity.[42–45] Additionally, the extracted quality factor (Q-factor) is found to be ≈7500, which is three times higher than the reported value of composite TMD and high-Q photonic cavity-based laser devices.[44–46] The high-Q value resembles a pure emission spectrum and robust frequency standard resulting from the coupled oscillation of photo-excited dipoles, both of which are characteristic features of SRE, where the previously designed optical cavity is not required.[1,2,9] Furthermore, the integrated photon intensity variation with pumping power density shows a power factor of 1.6 ± 0.1. Importantly, this is less than 2, the characteristic superlinearity factor for the SRE system of an atomic dipole ensemble.[1,9,11] The low value of the power factor can be rationalized by considering the absorption coefficient of the thin layers. The power factor is calculated against the pumping power density, assuming each pump photon contributes an excitonic dipole. Nevertheless, the extremely shallow thickness of the active medium associates a loss of the pumped photons; as a result, we observe the lesser value. The obtained output power density under the irradiation of 4 μW μm⁻² is found to be 4.2 × 10⁻¹² W per 100 × 100 μm².

In a conventional laser system, the optical cavity architecture requires a set of mirrors on either side of the gain medium. The optical emission from the gain medium then oscillates back and forth between these mirrors providing feedback to the emission and resulting in system lasing. While the cavity dimension determines the laser emission line in theory,[44–46] in practice, photons are generated and amplified in an optically lossy medium that induces spontaneous emission.[1,47] Superimposition of such spontaneous and stimulated lasing modes together with the thermal oscillation of the optical cavity cause a broadening of the linewidth of the lasing modes.[1,2,10] To obtain highly coherent laser modes, it is, therefore, essential to reduce the spontaneous emission caused by an optically lossy cavity and to stabilize the light amplification medium. In contrast, SREs are a promising alternative to circumvent such shortcomings of conventional lasers as the phase coherence in SREs is maintained by the collective oscillation of the dipoles, as discussed above.[1,2,5,13] Furthermore, unlike the conventional lasers, all emitted photons take part in the resultant generation of the emission lines, enhancing the laser’s efficiency.[1,1,9] Crucially, in the case of an SRE, in theory, only a few photons can stimulate coherent emission. This means SRE may not even exhibit a threshold power density value.[1,3,9] Thus, our observation of the superliner enhancement of output intensity in Figure 4B (inset) is consistent with the underlying principle of SRE.

2.5. The Critical Temperature for Coherent Exciton Coupling

The detailed temperature dependency of the output spectra at low temperature is shown in Figure 5A. Clearly, the sharp modes superimpose with the emission spectra below ≈15 K, the critical temperature (Tc) for the system to attain the superradiant emission. Note that the exciton population is proportional to the irradiation intensity. Concurrently, this also results in the laser heating of the sample. Thus, the influence of irradiation power density to obtain Tc cannot be eradicated. Similarly, the strength of the exciton coupling varies inversely with hBN thickness, and it is therefore also inversely related to the Tc (see Figure S8 in the Supporting Information).

2.6. Superradiance at Higher Pumping Intensity

A gradual enhancement of the laser power density to a value higher than ≈120 μW μm⁻² unveils another distinct feature of the heterostructure. As shown in Figure 5B, with the enhancement of the pumping power density beyond ≈120 μW μm⁻², the sharp peaks gradually decrease with the simultaneous enhancement of the FWHM. Such distinct dependence of superradiant emission intensity of radiatively coupled excitons in quantum-well structure has been reported previously.[49] In this study, the
gradual decrease of the sharp peak amplitude at higher power is again related to the emission from aggregated excitons. When the laser power is higher, the laser illumination induces phonons that fend the dipole-dipole synchronization. Additionally, the stronger pumping intensity results in photo-doping and the generation of the higher-order excitonic complex with different radiative recombination rates in the active TMD layers,[16,34,35,46] limiting the phase synchronization. The effect of trions and higher-order excitons on SRE has been separately discussed in Note S8 in the Supporting Information. Note that with the reduction of pumping energy, the sharp peaks reappear again. This feature of the power dependence also contrasts the SRE from previously observed sharp spikes in a WSe₂ layer under low temperature, where saturated quantum emissions from the layer are observed (see Notes S4 and S5 in the Supporting Information).[38–40,50]

2.7. Dependence of SRE Spectra on the hBN Spacer Thickness

We further studied the impact of the hBN spacer thickness on the intriguing behavior of the emission spectra, depicted in Figure 5C. It can be seen that as the hBN spacer thickness decreases, the sharpness of the emission gradually enhances. In addition, the peaks are redshifted as shown by the grey arrow in Figure 5C. For devices with an hBN thickness of ≈7 nm and higher, the spectrum barely indicates the appearance of the sharp modes. This result clearly shows that the interlayer exciton coupling plays an essential role in the generation of superradiant emission. Additionally, as a control experiment, we performed the spatially resolved PL measurement on hBN/WSe₂(top) (see Figure 1B) of the same sample under the same experimental parameters. The spatial average (1 μm × 1 μm) of the obtained spectra is shown in Figure S10 in the Supporting Information, where no pronounced sharp lines at ≈760 nm, similar to the emission at the heterojunction is obtained. These observations produce a stark contrast with the nature of previously reported observation of sharp emission lines in monolayer TMDs sandwiched by few-layered hBN.[28,51,52] However, the well-known Förster-type energy transfer exhibit a similar behavior as the separation between the donor and the acceptor varies.[53,54] We have compared our observation with resonant energy transfer processes in 2D systems in Note S9 in the Supporting Information. It is noted that such resonant energy transfer process at a similar experimental condition does not show any sharp emission.

2.8. The Underlying Mechanism

According to all the measurements described above, we now discuss the interlayer exciton coupling mechanism responsible for the ultralow threshold SRE. The interaction between interlayer excitons produces a weak van der Waals force, which attracts the coupled exciton towards the hBN interface. The excitons on both sides of the hBN spacer layer possess different polarities attached to the interface (see Figure 2C).[57] When the number of excitons is large enough, in order to minimize the total energy of the exciton ensemble, the excitons will self-assemble into an ordered array in a head-to-tail manner. This arrangement is similar to the J-aggregate, which occurs in organic molecules.[55] Under this condition, the
broad optical spectrum will become very narrow. Similarly, well-ordered excitons whichaggregate along the 2D plane will generate a coherent coupling and induce the SRE.\cite{12,36,37,55,56} All of the above experimental results can be well interpreted based on our proposed mechanism. The sensitive temperature dependence of the observed SRE occurs as the weak van der Waals attractive force is easily broken by small thermal energy, destroying the ordered array of the coupled coherent exciton. Similarly, the disappearance of SRE under high excitation power density can also be well understood. The requirement of the hBN spacer and the impact of the layer’s thickness can also be realized. The coherent behavior of aggregated excitons observed here possesses several unique features similar to the predictions given by many theoretical works on exciton condensates in bilayer TMDs.\cite{57,58} However, in the previous theoretical studies of exciton condensates, the conduction band minimum of one of the 2D layers must be adjusted to overlap with the valence band maximum of the other 2D layer in order to create an electron-hole droplet. In stark contrast, this rigid requirement has been overcome in our study.

Note that screening from the chalcogen containing sheets helps to confine the excitonic dipoles in the TMD monolayer. This effect results in overall suppression of environmental influences (i.e., interaction with encapsulating material with a different dielectric constant); on the other hand, the suppression of the dipole extension in the vertical direction catalyses the sensitivity of the inter-layer exciton coupling on the inter-layer distance.\cite{59}

Let us now provide a more detailed theoretical background of our observation. The WSe2/hBN/WSe2 heterostructure resembles a system of identical monolayers quantum wells stack containing excitons of similar radiative transition frequency and transition dipoles, where the separation between the monolayers in the stack is much lower than the wavelength of the emitted photons. The self-assembled highly ordered resonant excitons on surfaces of the hBN at low temperature produce a quasi-2D exciton reservoir where an array of N 2-level excitons oscillates in a common photon field in area A. Using Equation S1 in the Supporting Information, the dipole-dipole coupling \( J_{\mu\nu} \) between excitons in such 2D array can be expressed as Equation (1)\cite{60,61}

\[
J_{\mu\nu} = \frac{\sqrt{\pi} A}{2} \int_0^\infty \frac{d\theta}{2} \left[ \frac{1}{2} - \left( \mathbf{q} \cdot \mathbf{P} \right)^2 \right] e^{i\theta} \exp \left[ i(\omega - \omega') \right] \]

where \( r_{\mu\nu} = r_\mu - r_\nu \), and \( \mathbf{P} \) represents the polarization of the excitons, \( \mathbf{q} \) contains polarization angle \( \theta \) on the \( x-y \) plane, and \( \mathbf{P} \) is the principal value of the integral. With the analytical calculation, as shown in Note S10 in the Supporting Information, Equation (1) can be reduced to Equation (2) as follows

\[
J_{\mu\nu} = J_{2D} \left[ f(\xi) + ig(\xi) \right]
\]

where

\[
f(\xi) = 2 \left[ J_0(\xi) - \frac{J_1(\xi)}{\xi} + \left( \mathbf{p} \cdot \mathbf{r}_{\mu\nu} \right)^2 J_1(\xi) \right]
\]

\[
g(\xi) = 2Y_0(\xi) - 2 \left( \frac{Y_1(\xi)}{\xi} + 2 \left( \mathbf{p} \cdot \mathbf{r}_{\mu\nu} \right)^2 Y_1(\xi) \right) - \frac{4\xi^2}{\xi^2 - 1} \left[ 1 - 2 \left( \mathbf{p} \cdot \mathbf{r}_{\mu\nu} \right)^2 \right]
\]

in which, \( J_0(\xi) \) and \( Y_0(\xi) \) are the Bessel functions of the first and second kind, respectively. The functions \( f(\xi) \) and \( g(\xi) \) represent the coherent and dissipative coupling, which physically signifies the frequency shifts and the collective decay rate of coupled excitons pair that also determines the linewidth of the emission. The dissipative coupling at the limit of \( \xi \ll 1 \) approach to unity, resembling a high decay rate and narrow emission linewidth, which is a signature of obtaining the Dicke’s regime. As the separation between the excitons in the top and bottom TMD layers is much smaller than the emission wavelength, the system acquires the Dicke’s regime at low temperature, and the layer sensitive emission enhancement has been observed. Interestingly, in 2D coherent dipole-dipole interaction, both the orthogonal polarization \( (\mathbf{p} || \mathbf{r}_{\mu\nu}) \) and \( (\mathbf{p} \perp \mathbf{r}_{\mu\nu}) \) of light show the existence of Dicke’s regime. This is also consistent with the experimental setup where both the excitation and collection of the emitted photons are orthogonal to the TMD layers and the exciton dipoles. The coherent part of this limit show \( 2\pi (\xi^2 + \frac{1}{\xi^2}) \) asymptotic nature resembling a red-shift of the emitted frequency with decreasing values of \( \xi \). These observations are nicely consistent with the experimentally obtained results, as discussed before. Concurrently, the results are also consistent with the theoretical predictions of exciton superradiance in multiple quantum well stack system by Jasper Knoester.\cite{62}

Additionally, the concept of a superradiance relies on the build-up of a collective Dicke state out of the discrete states of N identical emitters. This scenario is meaningful for atomic states or spatially localized states due to 3D carrier confinement. Free excitons dominate the emission in monolayer TMDs. Thus, without additional confinement, one cannot expect a build-up of Dicke states due to interference between the electronic states. As a result, SRE is not expected to originate from the TMD monolayer sandwiched by hBN layers. In our design, we emphasize that the van der Waals attraction between the dipoles in either side of the hBN layer induces 3D confined aggregated excitons, which assists in build-up collective Dicke states in the WSe2/hBN/WSe2 heterostructure.

2.9. Selection of the Modes

At low temperatures, several sharp peaks appeared in the emission spectra. Interestingly, only a few of them dominate over the other and based on the hBN thickness the predominant emission peaks shift. The dominant peaks distribution has been provided in Figure 3B, where p1 is in the thinner (1.8 nm)
hBN layer, and p2 is at the thicker (6.6 nm) hBN layer. However, because of existing impurities and wrinklings, the spectra often vary even on a heterostructure of particular hBN thickness due to the obvious reason. To understand the consistency and stability of the obtained spectra, we conducted a series of single-point measurements on the 1.8 nm hBN layer over a range of temperatures for an extended period (Figure 5A). Clearly, we have not observed any randomness in terms of emitted modes or peak positions. Similar behavior has been obtained in Figures 4B and 5B. The selectivity of the emitted mode is contingent on the coherent coupling of a dipole with the neighboring dipoles. In the WSe2/hBN/WSe2 device, both WSe2 layers share a different dielectric environment; the bottom layer contacts SiO2 and hBN, and the top layer shares interfaces with hBN and air. This induces a slight variation of the exciton binding energy. Finally, those set of excitons that satisfy this condition ($\xi \ll 1$) couple coherently and produce sharp emissions. On the other hand, with the lateral resolution of our confocal measurement setup of $\approx 500$ nm, it is expected that a set of coherently coupled dipoles of varying energy over the different regions under the simultaneous illumination in the laser spot may satisfy $\xi \ll 1$. As a result, the emission spectra show the superimposition of a set of sharp peaks. At this point, it is not very straightforward to label the sharp peaks based on their origin. This work is aimed to demonstrate a proof-of-concept experiment, and more detailed investigations beyond the scope of the current work are necessary to label the basis of the sharp peaks carefully.

2.10. Interlayer Coupling Induced Change of Exciton Decay Dynamics

We have measured the exciton relaxation dynamics across the heterostructure and 1L-WSe2 using a time-resolved differential reflectivity ($dR/R$) measurement to further support our conclusions. A detailed description of the experimental setup and measurement conditions is provided in the Materials and Methods section. The observed decay kinetics at 1L-WSe2 (position C in Figure 6A, recorded at 725 nm at 10 K) shows a bi-exponential decay profile with fast and slow lifetimes of $\approx 10 – 20$ ps and $60 – 80$ ps, which are consistent with the previously published reports.$^{[63–66]}$ Interestingly, the decay kinetics changes dramatically at the heterostructure where a new fast decay channel appears – the corresponding characteristic time constant is $\approx 1.5 – 2$ ps for the heterostructure with 1.8 and 6.6 nm hBN spacer layer, respectively (see Figure 6A, positions A and B). The neutral and charge excitonic emission is separated at 10 K, as shown in Figure S12 in the Supporting Information. A comparison of neutral exciton and trion decay dynamic is provided in Figure S13A in the Supporting Information. Clearly, this extremely fast decay of the excitons at the heterostructure is due to the coupling of excitons, which causes the appearance of sharp peak at 725 nm (see Figure S12 in the Supporting Information, marked by the arrow). The emission time constants for the sharp peaks at 750–760 nm were found to be too short (sub-picosecond) to be analysed from the obtained transient $dR/R$ values as shown in

Figure 6. Coupled exciton lifetime obtained from time-resolved differential reflectivity ($dR/R$) measurements. A) Normalized exciton decay dynamics measured by degenerate pump-probe experiment at 10 K using 725 nm pump and probe pulses with average power 0.5 and 0.2 $\mu$W $\mu$m$^{-2}$, respectively. The dotted lines are the fitted curves using bi- and tri-exponential decay functions; the corresponding residuals are shown in the lower part of the figure. The marks A, B and C in the inset depict the corresponding positions on the sample where the signal was measured. The emission spectra recorded at the marked areas are shown in Figure S7 in the Supporting Information. B) Transient decay dynamics measured at thinner heterostructure (position A) at 10 K (using pulses at 725 nm) and 300 K (using pulses at 750 nm). The transient decay profile shows bi-exponential nature at 300 K, while it is tri-exponential with a faster lifetime component at 10 K.
Figure S13A in the Supporting Information, which are strikingly different from the time constant ($\approx$ ns) of bound excitons due to lattice impurities and Moiré excitons that occur in the similar energy ranges. [38–40,63,67,68] As the emitted photon intensity is inversely proportional to the radiative recombination rate, the observed shortening of lifetime fits well with the underlying emission mechanism. [69,70] This fast decay component is absent at higher temperatures in the measured dynamics (see Figure 6B and Figure S13B (Supporting Information)) because lattice phonons destroy the coherence between excitons. Moreover, the spectrally overlap of the emission from different emissive channels remains unresolved by differential reflectivity measurement.[65,71]

2.11. The Versatility of the Design: SRE from WS$_2$/hBN/WS$_2$ Heterostructure

We now demonstrate the versatility and robustness of our design by using the heterostructure based on different 2D materials, such as 1L WS$_2$, MoS$_2$, and few-layer hBN. The optical microscopy image of the heterostructure is depicted in Figure 7A. Interestingly, at low temperature, the emission spectra from the heterostructure also show the appearance of sharp peaks, as seen in Figure 7B. The distribution of the sharp peaks over the heterojunction is mapped in Figure 7C. This intriguing feature of emission spectra can be understood in terms of the aggregated dipoles in the WS$_2$ layers, as discussed above.

We performed second-order correlation function $g^{(2)}$ measurement using the Hanbury Brown and Twiss-type interferometer to obtain the emission correlation.$^{[72]}$ A detailed discussion of the measurement setup is included in the Materials and Methods section. We measured the temporal intensity correlation function at room temperature for the WS$_2$ based heterostructures due to the instrumental limitation. As shown in Figure 7D, the intensity correlation histograms for hBN thickness of 4.2 and 6.0 nm clearly reveal the bunching effect. Interestingly, the bunching effect is more pronounced in the heterostructure with a thinner hBN layer. Meanwhile, as expected, in monolayers both on SiO$_2$ and on hBN no correlated emission was obtained. This observation suggests that correlated emission occurs even at room temperature. With the reduction of the phonon population at low temperature, it is expected to have much robust correlated emission. Note that superradiant emission exhibits a giant bunching effect due to highly synchronized emission.$^{[73,74]}$ Thus, this experiment supports our the proposed mechanism of obtaining superradiant emission. The observation of such correlated emission from MoS$_2$ based heterostructure, as shown in Figure S14 in the Supporting Information, resembles the versatility of our design, suggesting the possibility of designing entangled quantum emitting devices by merely changing the TMD layers. A more detailed discussion on the observed
phenomena from the WS$_2$ and MoS$_2$ based devices will be reported separately.

3. Conclusion

In conclusion, we have successfully designed and demonstrated van der Waals heterostructure-based novel SRE devices, where the collective oscillation of excitonic dipoles produces a continuous and cumulative emission with a high degree of phase coherence. In principle, the as-designed SRE device can potentially address multiple fundamental challenges encountered by the conventional lasers; for instance, obtained ultranarrow optical transitions of ≈ 0.1 nm are more spectrally pure. The corresponding coherence length is found to be comparable with state-of-the-art laser systems, and Q-factor is much higher than all previously reported TMD-based lasers. These intriguing properties can dramatically enhance the performance of advanced atomic clocks and associated technologies, such as encryption, quantum communications, GPS navigation systems, astronomical instruments, etc. Notably, the synchronization of the dipole oscillation is very sensitive to the presence of phonons, which degrade the coupling and the phase coherence of the emitted photons due to weak binding of van der Waals force. We have also supported our experimental results with the theoretical model. The simple SRE device presented here is based on the emerging 2D materials, and we suggest this can be extended to many other materials systems. We believe these results provide new avenues for the realization of stable, high-performance, ultranarrow linewidth coherent light sources with a high spectral tunability and a very simple fabrication process.

4. Experimental Section/Methods

Commercially purchased (2D Semiconductors) bulk TMDs and hBN layer were exfoliated on thin PDMS membrane with a standard exfoliation technique. These were then transferred onto a 300 nm SiO$_2$/Si substrate to obtain the final heterostructure. OM images were captured using an Olympus microscope. AFM measurements were carried out in tapping mode with a Dimension Icon microscope (Bruker Inc.). The images were captured using Bruker SCANASYST-AIR probes (k = 0.4 N/m, f$_p$ = 70 kHz, nominal tip radius = 2 nm). The obtained topography was processed using the Gwyddion software.

Spectroscopic Characterization: Temperature-dependent PL spectra were measured using a low temperature confocal Raman microscope (attoRAMAN, attocube) inserted in a Physical Property Measurement System (PPMS, Quantum Design). Prior to measurement, the sample space was flushed with He gas, and the chamber was evacuated to a pressure of 5 mbar. The PL spectra were recorded using a WITec Alpha300 Raman microscope consisting of 1X71 body (Olympus, Japan). The excitation light emitted from a pulsed diode laser (PicoTa, 532 nm, Toptica, Germany) with 40 MHz repetition rate was coupled to a polarization-maintaining single-mode optical fiber and at the output collimated with an air space objective (UPLSAPO 4X, Olympus, Japan) with 543 dichroic mirror (ZT543rdcxt, Chroma, USA). Laser intensity at the back aperture of the objective was adjusted to 100 nW. The collected emission signal was then split between two single-photon avalanche diodes (PDM series, MPO, Italy) using 50/50 beam splitter plate. Two emission filters (605/50, Chroma, USA) were placed in front of each detector to minimize the effect of the detector afterglow. The autocorrelation functions were calculated by cross-correlating the signal from the two detectors with home-written scripts in Matlab (Mathworks, USA).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Research data are not shared.
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