
Supplementary information

Chiral superfluorescence from perovskite superlattices at room temperature

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Supplementary Information for

Chiral superfluorescence from perovskite superlattices at room temperature

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Supplementary Note 1: Characteristics of superfluorescence

1.1 Overview of superfluorescence

Superfluorescence (SF) is a fundamental quantum optical phenomenon characterized by the spontaneous emergence of macroscopic quantum coherence from an initially incoherent ensemble of emitters. In this process, a collection of inverted two-level systems, initially prepared without phase correlation, develops collective coherence through resonant exchange of virtual photons mediated by quantum fluctuations of the electromagnetic vacuum.

The conceptual foundation for cooperative radiation was first established by Dicke in 1954, who introduced the term "superradiance" to describe enhanced emission from N identical atoms sharing a collective excited state with a pre-existing macroscopic dipole¹. In contrast, SF emerges without initial dipole alignment, relying instead on self-organization through inter-emitter coupling. The theoretical framework for SF in systems smaller than the emission wavelength was first developed by Eberly and Rehler^{2,3} in 1969, with subsequent quantum mechanical treatments by Bonifacio (1975) and Haake (1979), among others, providing rigorous analytical tools for describing more general systems⁴⁻⁹. Through these studies, the temporal profile of a pure superfluorescent pulse (in the absence of dephasing) was determined as

$$I_{\text{SF}}(t) \propto N^2 \text{sech}^2\left(\frac{t - \tau_D}{\tau_P}\right) \quad (1)$$

where N is the number of excited emitters, $\tau_D = (1/\Gamma)\ln(N)/(N + 1)$ is the delay time, and $\tau_P = 2\tau_D/\ln(N)$ is the pulse width. These scaling laws allow SF to be distinguished from other, incoherent radiation processes.

1.2 Burnham-Chiao ringing in superfluorescence

The temporal profile given in Eq. (1) is applicable for a single superfluorescent burst in the limit where the propagation of photons within the active sample is negligible. In optically thick media, this condition can be violated, leading to periodic re-absorption and re-emission of coherent light. These processes manifest as damped oscillations in the temporal SF profile, known as Burnham-Chiao ringing^{9,10}. Burnham-Chiao ringing is an inherently non-Markovian phenomenon, for which a concise analytical solution for the time evolution of the SF intensity does not exist. In practice, it is sufficient to describe the ringing behavior as a sum of individual superfluorescent pulses^{4,11},

$$I_{\text{SF}}(t) \propto \sum_i^4 N^2 \text{sech}^2 \left(\frac{t - \tau_{\text{Di}}}{\tau_{\text{pi}}} \right). \quad (2)$$

In Fig. 2b of the main text, we fit our data to a sum of four sech^2 functions at high excitation fluences to account for the ringing effect. Note that the corresponding linear-scale plots (Supplementary Fig. 17a, Extended Data Figs. 6a and 7a) show that the initial SF burst significantly outweighs subsequent oscillations, indicating that propagation-related re-excitation is weak.

Supplementary Note 2: Structure analysis of vertical-orientated superlattices

Grazing-incidence wide-angle X-ray scattering (GIWAXS) provides enhanced structural characterization compared to traditional powder diffraction by establishing a spatial reference frame through the thin film plane. In GIWAXS, each pixel is mapped onto a two-dimensional grid containing both in-plane (q_r) and out-of-plane (q_z) scattering vector components. When superlattice structures form in 2D ($n = 1$) and quasi-2D perovskites, the extended periodicity produces small-angle peaks ($q < 1 \text{ \AA}^{-1}$) corresponding to larger lattice spacings.

Our GIWAXS results (Extended Data Fig. 3) show clear streak patterns for the superlattices. Analysis of the q_r (in-plane, parallel to substrate surface) and q_z (out-of-plane) intensity distributions reveal distinct structural characteristics: the in-plane signal shows relatively uniform intensity distribution along the (0k0) directions at small angles, while the out-of-plane signal is dominated by a strong (111) peak. This pattern indicates homogeneous in-plane structure with vertically oriented multi-quantum wells perpendicular to the substrate surface.

Complementary atomic-resolution imaging using scanning transmission electron microscopy (STEM) with high-angle annular dark field (HAADF) detection confirms these structural findings. Since HAADF-STEM intensity scales approximately with the square of atomic number, it provides chemical composition information at atomic resolution. Our cross-sectional STEM images (Extended Data Fig. 2) reveal high-quality interface formation with perovskite quantum wells growing perpendicular to the MAPbBr₃ substrate. The measured quantum well width of $\sim 1.8 \text{ nm}$ corresponds to three $[\text{PbI}_6]^{4-}$ octahedral units, consistent with the $n=3$ structure of our superlattices.

Supplementary Note 3: Optical simulations and photonic effects in perovskite superlattices

A numerical model using the finite element method (FEM) was developed to describe photonic properties of the perovskite superlattices. The FEM was implemented using the Wave Optics Module in the COMSOL Multiphysics software. Scattering boundary conditions were set at each boundary and the perfect matching layer was set along the z direction. A solution for the electric field $E(\mathbf{r})$ was obtained and the absorption (A) was calculated as:

$$A(\omega) = \frac{\iiint_V 0.5\epsilon_i\omega|E(\mathbf{r})|^2 dV}{P_{in}}, \quad (3)$$

where V represents the overall volume of the region of interest, P_{in} is the incident light power, ω is the angular frequency of the incident light, ϵ_i is the imaginary part of the perovskite material refractive index, and $E(\mathbf{r})$ is the electric field at the position of (x, y, z) . The simulated superlattice structure was modeled as a hollow rectangular piped with a height of 3 μm , edge length of 1 μm , and wall thickness of 100 nm (see insets in Extended Data Figs. 4f,g). The simulated absorption spectra agree well with our measurements and are independent of the linear polarization direction of the pump laser (Extended Data Fig. 4).

We also investigated whether the 700 nm edge state emission could result from waveguiding of bulk excitons (emitting at 620 nm) to the superlattice periphery. Simulations were performed using the finite-difference time-domain (FDTD) method in Ansys Lumerical FDTD. Four linearly polarized (LP) dipole sources, with emission spectra based on the measured bulk exciton emission peaking at 620 nm (Extended Data Fig. 5g), were placed at the center of each rectangular layer to simulate isotropic bulk emission. A z -normal field monitor was placed at the top surface to record near-field propagation. The simulations (Extended Data Fig. 4i) show no enhancement of the 700 nm electric field at the edges, ruling out significant waveguiding effects. Therefore, the 700 nm luminescence observed at the edges of both exfoliated single crystal and superlattices (Extended Data Fig. 5) originates from intrinsic emissive states localized at the edges.

To evaluate photonic contributions to the observed circular polarization, we performed simulations of circularly polarized (CP) light absorption and far-field radiation from CP dipoles inside the superlattices (Extended Data Figs. 4g, 4j). For the simulation of CP light absorption, the LP incident wave set on the far-field port was replaced by two orthogonal LP waves with a phase difference of $\pm \pi/2$ in COMSOL Multiphysics. For simulating the far-field radiation

intensity of opposite handedness, two dipole sources with perpendicular polarization directions and a phase difference of $\pm \pi/2$ were positioned in the bulk center of the perovskite superlattices in Ansys Lumerical FDTD. The simulations reveal identical optical responses for LCP and RCP light, confirming that photonic effects do not account for the observed circularly polarized chiral SF.

Supplementary Note 4: Superfluorescence from edge states—evidence and implications

Our combined experimental and numerical results indicate that the SF emission at ~695–710 nm originates from the edge states of PEA/SMBA/RMBA quasi-2D perovskite superlattices. To confirm this origin, we grew quasi-2D single crystals of identical composition to our SMBA superlattices ((PEA)_{1.2}(SMBA)_{0.8}(MA)₂Pb₃I₁₀), and prepared exfoliated microplates for comparison. While the centers of these microplates show bulk exciton emission at 620 nm, their edges (tens of nanometers) emit at ~700 nm—directly matching the wavelength of the SF observed in our superlattices (Extended Data Fig. 3). In the superlattices themselves, the bulk exciton emission at ~600 nm appears only under very high fluence above the SF threshold (Supplementary Fig. 3) and scales linearly with pump power, in stark contrast to the superlinear scaling of the edge state SF emission.

Additionally, our measurements indicate that the excited dipole density N in the edge states follows a superlinear dependence on pump power ($I_{SE} \propto N \propto P^{1.5-1.6}$), a behavior characteristic of these specific edge states in quasi-2D perovskites¹². This intermediate scaling reflects the hybrid nature of the edge states in $n = 3$ structure, which lies between the purely excitonic ($I \propto P$) and bimolecular ($I \propto P^2$) regimes.

Furthermore, the significant spectral separation (~80 nm) and distinct lifetimes between edge state and bulk emissions (Supplementary Figs. 3, 4) rule out waveguiding of bulk emission as the origin of the edge luminescence. Consistent with previous works^{13,14}, our density functional theory (DFT) calculations further confirm the existence of edge states with a narrower bandgap (Supplementary Fig. 5), therefore leading to a funneling of excitation population down the energy gradient from the bulk to the edge.

Since thermal scattering is a primary source of decoherence at elevated temperatures, we examined the temperature dependence of the emission linewidth. The linewidth broadening of spontaneous emission from the edge states remains remarkably small (~10 meV) from 77 K to 300 K (Supplementary Fig. 6a), in stark contrast to the substantial broadening (~40 – 100 meV) observed for bulk excitons in quasi-2D perovskite microplates (Supplementary Fig. 6b and Ref.¹⁵), bimolecular emission in 3D perovskites¹⁶, or exciton emission in perovskite nanocrystals¹⁷. This indicates significantly suppressed phonon scattering in the edge states, enabling coherence retention. The superior optoelectronic properties of these edge states are further evidenced by their high photoluminescence quantum yield (PLQY) and long emission lifetimes (Supplementary Fig. 6c,d), suggesting effective protection against nonradiative decay

channels such as electron-phonon coupling and impurity scattering (see also Supplementary Note 8.4). Similar robust edge state emission has been reported in $\text{BA}_2\text{MA}_{n-1}\text{Pb}_n\text{I}_{3n+1}$ ($n \geq 3$) systems, where it also contributes to enhanced photovoltaic performance with long lifetime¹². Our transient absorption measurements further confirm that photoexcited carriers localize to these edge states within ~ 1 ps after excitation (Supplementary Fig. 6e), underscoring their role as the primary emitting species in the observed SF.

For our vertically-oriented superlattices, the exposed edges lie at the top of our perovskite samples, facing the direction of SF (the bottom edges are quenched by the growth substrate, Extended Data Fig. 2). This is consistent with the forward-backward asymmetry required to yield a nonzero circular polarization during chiral SF. As discussed in the main text, emergent spin-orbit coupling locks the polarization of the light to the quasimomentum (and group velocity) of the superfluorescent collective mode. Preferential propagation of quasi-localized wavepackets towards the top edge prior to emission is therefore predicted by our theory.

Supplementary Note 5: Coherent coupling of emitters in superlattices

SF requires the coherent coupling of individual quantum emitters through shared electromagnetic modes, eventually leading to an enhanced radiative decay rate. This coupling is only possible when many closely spaced emitters (relative to the emission wavelength) are excited with transition energy differences less than their radiative linewidths. In the main text, we present multiple lines of evidence for SF in our quasi-2D perovskite superlattices, which enable coherent coupling between properly aligned emitters. Here, we provide an additional, independent verification of coherent coupling through the enhanced ratio of photoluminescence (PL) decay rates between quasi-resonant and non-resonant excitations ($\tau_{qr}^{-1}/\tau_{nr}^{-1}$) (Ref 18). When probed at low pump fluence, this ratio peaks at the same wavelength where edge state SF emerges under high pump fluence in our achiral and chiral superlattices (Supplementary Figs. 7–9). This key observation can be interpreted as follows:

- Under non-resonant excitation (450 nm for our superlattices), dipole transitions are excited well above the emission energy. During the subsequent relaxation process, these dipoles lose their initial phase relationship and can transition to other non-resonant states. This significantly reduces the probability of finding multiple dipoles with transition energies matching the emission wavelength within the characteristic radiative coupling distance. Consequently, there is no collective enhancement to the PL decay rate under non-resonant conditions at low pump fluence.
- By contrast, the density of dipoles with closely spaced transition energies is highest near the peak emission wavelength. Therefore, quasi-resonant excitation at 650 nm results in a greater likelihood of exciting multiple dipoles within this spectral band. Consequently, these dipoles are able to interact collectively with the common radiation field, which manifests experimentally as a faster PL decay rate. The enhanced ratio of PL decay rates between quasi-resonant and non-resonant excitations is therefore direct evidence for coherent coupling between emitters. This measurement scheme was previously used to verify coherent coupling between individual quantum dots¹⁸, and our results extend these observations to the case of chiral perovskite superlattices.

It is important to clarify that the measurements described above at low pump fluence are perfectly consistent with the observation of SF under 550 nm excitation at high pump fluence. Whereas quasi-resonant, low power excitation at 650 nm prepares an initially correlated state leading to enhanced PL decay (*i.e.*, few-photon superradiance), collective enhancement via SF

is only observed under 550 nm excitation at high pump fluence. Under these conditions, the density of photoexcited dipoles is large enough to overcome the non-resonant nature of the initial excitation. This interpretation is also supported by our theoretical modeling, which shows that SF only takes place when the dephasing rate γ_ϕ is smaller than the density-dependent collective decay rate ΓN . As such, the independent observations of an enhanced PL decay rate at the ~ 700 nm SF wavelength under quasi-resonant excitation at low power together with the observation of SF under non-resonant excitation at high power provide complementary evidence for coherent coupling between dipoles in our perovskite superlattices. Finally, we note that control experiments performed on disordered polycrystalline MAPbI₃ films (containing multiple randomly oriented grain boundaries) show no such enhancement in the PL decay rate ratio (Supplementary Fig. 10), indicating that this effect is unique to the ordered nature of the superlattices.

Supplementary Note 6: Distinguishing amplified spontaneous emission from superfluorescence

A hallmark of amplified spontaneous emission (ASE) is its dependence on the optical path, as light amplification increases with propagation distance. To rule out ASE in our superlattices, we performed systematic directional emission measurements under a stripe-shaped excitation beam. In control experiments performed on CsPbBr₃ nanocrystal films, we observed the expected path length dependence of ASE (Supplementary Fig. 13): edge-collected emission was significantly stronger than forward-collected emission, consistent with ASE behavior reported previously^{19,20}. In stark contrast, our perovskite superlattices consistently exhibited stronger emission in the forward direction (normal to the sample surface), regardless of excitation geometry (stripe-shaped or point-beam excitation) (Supplementary Fig. 14). This clear and consistent departure from ASE-like directionality provides robust experimental evidence that the observed emission from our superlattices does not originate from ASE, thus supporting its identification as superfluorescence.

To further rule out the possibility that the enhanced forward emission originates from photonic effects within the crisscross superlattices, we performed numerical simulations to evaluate the emission intensity anisotropy. According to Lorentz reciprocity, the field amplitude radiated by a source into a given direction and polarization is proportional to the coupling between that source and a field of the same polarization incident in the opposite direction²¹. As such, the far-field emission anisotropy can be evaluated by computing the field components within the active medium under plane wave illumination. Accordingly, we compared the horizontal-to-vertical emission intensity ratio (I_h/I_v) for the crisscross superlattices with that of a bulk perovskite crystal with identical width, length, and height (Supplementary Fig. 13f) under equivalent incident power. Assuming a random distribution and orientation of dipole emitters, the ratio is derived from the volume-averaged electric field intensity within the active volume V as

$$\frac{I_h}{I_v} = \frac{(\int_V |\mathbf{E}_h(x,y,z)|^2 dV)}{(\int_V |\mathbf{E}_v(x,y,z)|^2 dV)}, \quad (4)$$

where $\mathbf{E}$ is the electric field. As shown in Supplementary Fig. 13g, the emission ratio is comparable between the superlattice and bulk models, irrespective of whether the integration encompasses the entire crystal or is confined to the top 50 nm. This indicates that the crisscross network does not preferentially scatter the emitted light out-of-plane. Thus, the forward-directed emission observed in our superlattices cannot be ascribed to photonic effects or ASE.

Supplementary Note 7: Spatial coherence of superfluorescence

To characterize the spatial coherence of the superfluorescent light emitted from our perovskite superlattices, we recorded the interference patterns using a Michelson interferometer. The Michelson interferometer produces interference fringes by splitting a light beam into two parts and then recombining them after they have traveled different optical paths; one beam is reflected towards mirror 1, while the other is transmitted towards mirror 2. The mirrors then reflect the beams back toward the beam splitter and the CCD camera (Supplementary Fig. 15). Since the phases of the incident superfluorescent beams are highly correlated, the interference pattern of dark and bright fringes can be seen on the CCD camera. In the case of a spontaneous emission (i.e., not superfluorescent) source, no interference signatures were observed, excluding the possibility that the fringes originated from the excitation laser pulses.

The interference fringes recorded by the Michelson interferometer show evidence of spatial coherences up to ~ 1 mm in length. Importantly, this length scale, which is indicative of correlated emission within a macroscopic ensemble of coupled SF domains, is distinct from the temporal coherence length of an individual SF domain and also depends on factors such as the size and uniformity of the light source. The temporal coherence length (L_C) can be calculated using the formula $L_C \approx \frac{\lambda^2}{\Delta\lambda}$, where λ is the central wavelength of the emitted light and $\Delta\lambda$ is the spectral FWHM linewidth²². Based on the data from Fig. 1f of the main text, the coherence length in the SMBA perovskite is approximately $L_C \approx 50 \mu\text{m}$ at $172 \mu\text{J}/\text{cm}^2$. This is a conservative estimate as the SF linewidth may be artificially broadened by the excitation at 550 nm and subsequent relaxation (Supplementary Note 8.3). Crucially, the measured temporal coherence length is significantly larger than the spacing between individual emitters in our superlattices, thus allowing them to couple coherently into collective domains. The highly uniform and large-area nature of our superlattices enables the collective emission from this vast array of coupled domains to exhibit extended spatial coherence over millimeter scales.

Supplementary Note 8: Measurements and explanations for long room-temperature coherence times

8.1 Estimating coherence times for room-temperature superfluorescence

In optical systems comprised of multiple emitters, broadening of the transition linewidth generally contains both homogeneous and inhomogeneous contributions. Homogeneous broadening arises from interactions that affect all emitters identically, such as radiative and nonradiative recombination or coupling to thermal lattice vibrations. For a purely homogeneously broadened transition, the dipole coherence decays exponentially with characteristic time²³

$$T_2 = \left(\frac{1}{2T_1} + \frac{1}{T_\phi} \right)^{-1}, \quad (5)$$

where T_1 is the population decay time and T_ϕ is the pure dephasing time. By contrast, inhomogeneous broadening results from interactions that affect the emitters differently (e.g., Doppler broadening in atomic gases or sample inhomogeneities in the solid-state), leading to a distribution of resonance frequencies. At room-temperature, PL in hybrid perovskites typically exhibits both homogeneous and inhomogeneous broadening²⁴. In this case, the dipole coherence does not follow a purely exponential decay and is instead described by an effective coherence time T_2^* . Because T_2^* is necessarily shorter than T_2 (homogeneous broadening is always present), T_2^* provides a lower bound on T_2 .

For SF to occur, the ensemble of dipoles must retain coherence long enough to spontaneously synchronize and emit a collective burst. This requires that the SF buildup (delay) time τ_D be shorter than the effective dephasing time T_2^* (Ref. 4). As such, τ_D measured near the SF threshold provides a conservative lower limit on T_2^* . In our SMBA superlattice sample, the delay time near threshold is $\tau_D = 11.6$ ps (Fig. 1c), which is comparable to the values of $\tau_D = 12.5$ ps and 23 ps measured at room temperature and 78 K, respectively, in quasi-2D PEA:CsPbBr₃ hybrid perovskite films exhibiting SF^{11,25}.

8.2 Four-wave mixing methodology and T_2 extraction

To directly verify the long coherence times in our samples, we performed an independent measurement of the homogeneous dephasing time T_2 using time-integrated degenerate two-pulse four-wave mixing (FWM). In this experiment, two non-collinear ultrafast laser pulses with frequency ω and wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ are incident on the sample at times t_1 and t_2 ,

respectively (Supplementary Fig. 18a,b). For ω on resonance with the dipole transition, the nonlinear interaction generates a third-order polarization within the medium that radiates a signal in the phase-matched direction $\mathbf{k}_s = 2\mathbf{k}_2 - \mathbf{k}_1$. The intensity of this signal $I_s(\tau_{12})$ is then recorded as a function of the variable time delay $\tau_{12} = t_2 - t_1$. In the limit where the pulse widths are much shorter than the T_1 and T_2 times of the optical transition, the FWM signal serves as a direct probe of T_2 (Ref²⁶).

Importantly, the interpretation of the FWM intensity depends on whether the dipole transition is dominated by homogeneous or inhomogeneous broadening. For a Gaussian distribution of resonance frequencies with standard deviation $\delta\omega$, the intensity of the FWM signal in the $\mathbf{k}_s$ direction is given in the inhomogeneous limit ($\delta\omega \gg 1/T_2$) by²⁶

$$I_s(\tau_{12}) \propto \exp\left(-\frac{4\tau_{12}}{T_2}\right) \left[1 + \operatorname{erf}\left(\sqrt{\frac{2}{\pi}} \delta\omega \tau_{12}\right)\right], \quad (6)$$

where $\operatorname{erf}(x) = (2/\sqrt{\pi}) \int_0^x \exp(-t^2) dt$ is the error function. By contrast, for a homogeneously broadened transition ($\delta\omega \approx 0$), the signal intensity is instead described by the simpler form²⁶

$$I_s(\tau_{12}) \propto \exp\left(-\frac{2\tau_{12}}{T_2}\right). \quad (7)$$

The signal intensity peaks at $\tau_{12} > 0$ and $\tau_{12} = 0$ in the inhomogeneous and homogenous limits, respectively, allowing the appropriate model to be identified directly from the time delay measurements.

In our experiments, we directly probe the 700 nm transition of the superfluorescent edge states using 50 fs laser pulses. At all pump fluences studied, the signal intensity peaks at $\tau_{12} = 0$ and exhibits a straight-line dependence on a semi-log plot (Supplementary Fig. 18c-e). This confirms a mono-exponential decay of the macroscopic polarization, consistent with Eq. (7) above for a homogeneously broadened transition. The measured T_2 decreases monotonically with increasing pump fluence (from 72 ± 5 ps to 11 ± 3 ps for the SMBA superlattices as the 700 nm pump fluence increases from 25 to $380 \mu\text{J cm}^{-2}$). This reduction is attributed to excitation-induced dephasing mechanisms, such as enhanced carrier-carrier scattering at higher densities. Since in our materials the dephasing time T_2 is much shorter than the population decay time T_1 (Supplementary Fig. 6c), the measured T_2 thus provides a good approximation for the pure dephasing time T_ϕ .

Although, a direct quantitative comparison between the homogeneous T_2 extracted from FWM and the effective T_2^* inferred from SF delay measurements under 550 nm excitation is not straightforward, the trend towards $T_2 \approx T_2^*$ at very high pump fluence suggests a dominant contribution from pure dephasing. At lower pump fluences closer to the SF threshold, the larger values of T_2 compared to $T_2^* \gtrsim \tau_D$ may indicate a contribution from inhomogeneous broadening. This likely stems from the nonspecific recruitment of dipoles with a distribution of resonance frequencies during the initial 550 nm excitation and subsequent relaxation, as compared to the spectrally narrow probe at 700 nm employed during FWM (see also Supplementary Note 8.3 below).

8.3 Reconciling broad emission linewidths with long coherence times

For an isolated two-level system, the emission linewidth is directly related to the dipole coherence time through the Fourier transform of the dipole correlation function. In our experiments, however, the situation is more complex. Excitation is performed at 550 nm, while emission occurs near 700 nm, implying the involvement of multiple electronic transitions and relaxation pathways, potentially including vibronic contributions. Consequently, the PL FWHM does not correspond to a single optical transition and cannot be directly used to infer the coherence time of the edge states.

Compared with linear spectroscopy, the non-linear FWM signal enables a more direct assessment of coherent dynamics²⁶. At high pump fluence (Fig. 1f), we observe a significant narrowing of the SF linewidth ($\Delta\lambda_{\text{SF}} \approx 7$ nm FWHM) compared to the broad spontaneous-emission PL linewidth at low fluence ($\Delta\lambda_{\text{PL}} \approx 50$ nm FWHM). This signature further indicates that the PL spectra are comprised of multiple transition energies, only some of which experience SF. Similar spectral narrowing accompanying SF has been reported in Refs. 11 and 25. These observations indicate that, although the macroscopic sample is strongly inhomogeneously broadened, SF arises from a more homogeneous sub-ensemble that retains coherence over longer timescales²⁷.

Even at high pump fluence, the SF linewidth remains broader than the $\Delta\lambda \approx \frac{\lambda^2}{\pi c T_2^*}$ expected for a single SF domain. This observation is also consistent with previous studies of room-temperature SF in hybrid perovskites (Refs. 11 and 25). We attribute this residual broadening to inhomogeneities among independent SF domains, emission from which collectively contributes to the ensemble SF spectrum following excitation and relaxation. Whereas the

longer T_2^* estimated from the delay time dynamics reflects the relative homogeneity of individual domains, the broader overall SF linewidth implies simultaneous emission from multiple domains across the macroscopic sample (Supplementary Note 7). As such, the resulting hierarchy of linewidths, $\Delta\lambda_{\text{PL}} > \Delta\lambda_{\text{SF}} > \Delta\lambda_{T_2^*} \gtrsim \Delta\lambda_{T_2}$ (where T_2^* and T_2 refer to individual SF domains) is consistent with both prior reports^{11,25} and our independent measurements.

8.4 Physical origin of the long coherence time

As discussed above, our T_2^* times inferred from the SF delay time τ_D near the SF threshold are consistent with recent reports of 78 K and room-temperature SF in quasi-2D hybrid perovskite thin films^{11,25}. Given the close similarities between our observations and those in the literature (including the material class, comparable T_2^* values, and SF-associated spectral narrowing), we suspect that related physical mechanisms, such as polaron dressing and soliton formation^{11,25}, may also contribute to the long coherence times in our samples.

At the same time, there are differences between our observations and those reported for PEA:CsPbBr₃ films^{11,25}. In particular, SF in our superlattices occurs at a longer wavelength (~ 700 nm) and with a higher threshold pump fluence, possibly due to weaker radiative coupling at longer wavelengths, which necessitates a higher dipole density to reach the SF threshold. The fact that we observe SF under these conditions therefore underscores the presence of sufficiently long coherence times in the SF-participating states, even at elevated excitation densities. Correspondingly, the T_2 times measured in our superlattices are longer than those measured in our quasi-2D (PEA)_{1.2}(SMBA)_{0.8}(MA)_{*n*-1}Pb_{*n*}I_{3*n*+1} Ruddlesden–Popper (RP) perovskite thin film control sample (Supplementary Fig. 18), which contains primarily bulk excitons and exhibits no SF at room temperature (Supplementary Fig. 25). This contrast suggests that the superlattice structure offers additional protection against decoherence. We attribute this enhanced protection to the formation of edge states within the superlattices (Supplementary Note 4).

Several complementary measurements suggest that edge states in hybrid perovskites may be less susceptible to decoherence than bulk excitons. First, the increase in PL linewidth from 77 K to room-temperature is significantly smaller for the superlattice edge states (~ 10 meV) compared with the bulk exciton broadening (~ 39 meV) in the exfoliated $n = 3$ microplate (Supplementary Fig. 6a,b; Supplementary Note 4). This weaker temperature-dependent broadening indicates that the edge states are less sensitive to phonon-induced dephasing.

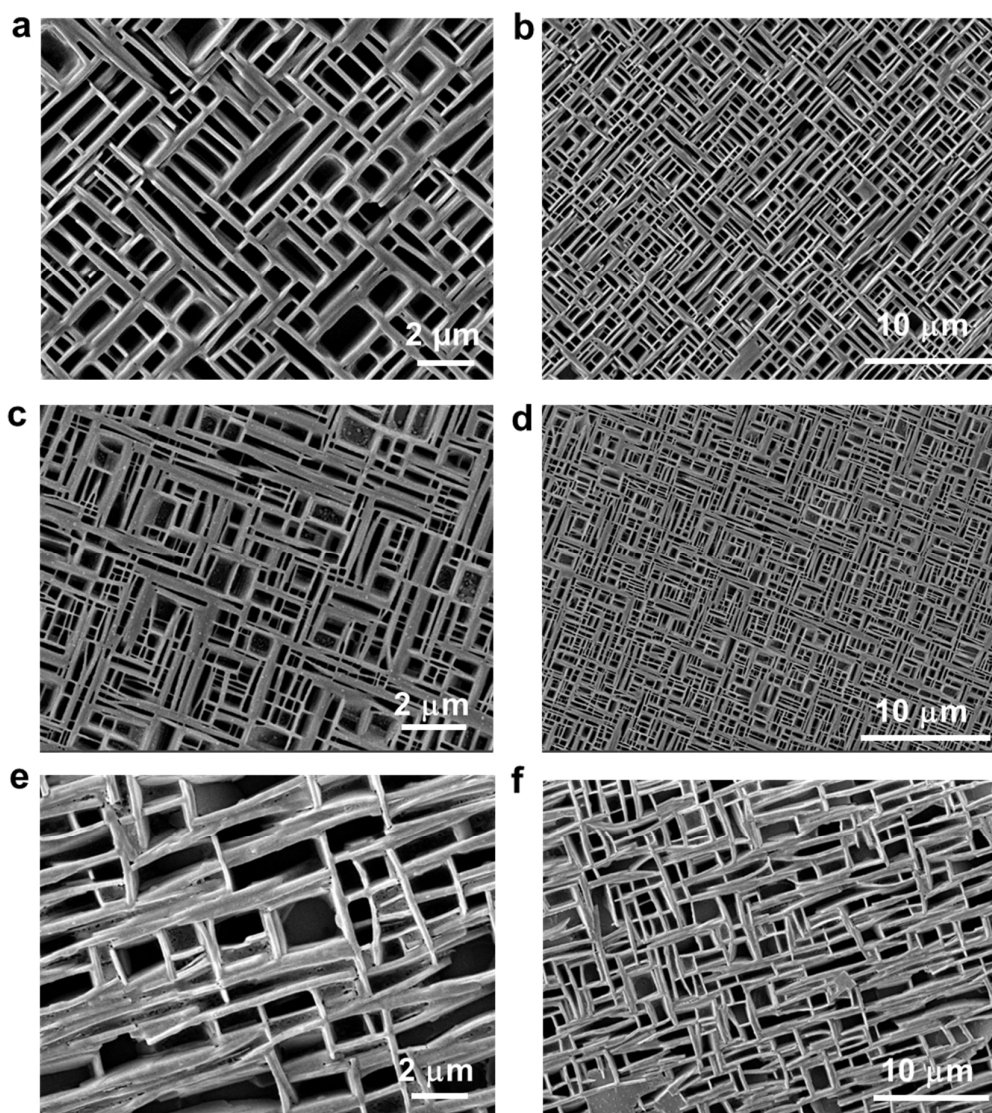
Second, the radiative lifetime of the edge states (1.5 ns in the superlattices, 1.3 ns in the exfoliated microplate) is substantially longer than the lifetime of bulk excitons (0.32 ns in the exfoliated microplate) (Supplementary Fig. 6c). This extended lifetime is accompanied by a significantly larger PLQY in the superlattices (Supplementary Fig. 6d). These measurements are consistent with Ref. 12, which showed that edge state localization in similar quasi-2D $(\text{BA})_2(\text{MA})_{n-1}\text{Pb}_n\text{I}_{3n+1}$ ($n \geq 3$) hybrid perovskite films significantly enhanced PL lifetimes and PLQY at room-temperature. While excited state lifetime and PLQY do not directly quantify coherence, these results indicate that the localized edge states experience a substantially weaker coupling to their environment, resulting in reductions in dephasing and nonradiative relaxation.

Consistent with this interpretation, multiple studies have experimentally and theoretically shown that exciton localization can reduce electron-phonon coupling, thereby leading to reduced inelastic scattering and extended coherence time²⁸⁻³². For instance, Ref. 32 also concluded that localization in similar perovskite systems contributed to a longer T_2 time by mediating a reduction in phonon-assisted relaxation. These effects may be especially pronounced in our samples, where ultrafast (sub-picosecond) trapping of carriers from the bulk into deep edge states (Supplementary Note 4, Supplementary Fig. 6f) reduces time spent in the more rapidly dephased bulk. The energetic separation between the bulk exciton resonance at 2.0 eV and the edge state energy of 1.77 eV renders thermal activation back into the bulk continuum negligible at room temperature. As a result, the edge states remain confined within the localized manifold, where inelastic scattering channels are strongly suppressed. In aggregate, these processes likely explain the difference in T_2 times between our superlattices and our thin film control sample, and together with the additional polaron dressing described in Refs. 11 and 25, provide a plausible explanation for the long coherence times overall.

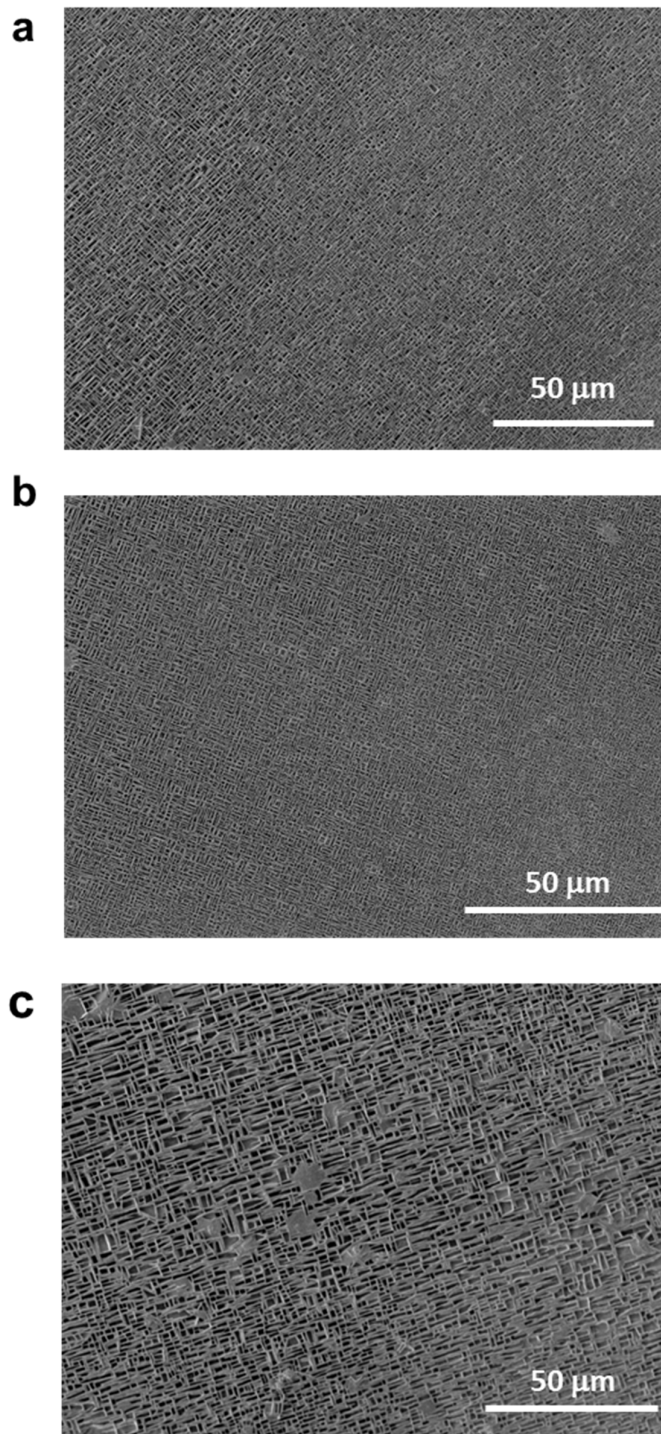
Supplementary Note 9: Chirality transfer from the chiral ligands to the perovskite superlattices

Supplementary Fig. 20 shows the measured circular dichroism (CD) spectra of achiral PEA, chiral SMBA, and chiral RMBA superlattices, with the ~ 700 nm peaks aligning with the wavelength of edge state emission. The chiral perovskites crystallize as 2D hybrid organic-inorganic structures, featuring anionic $[\text{PbI}_4]^{2-}$ layers composed of corner-sharing PbI_6 octahedra that are interspersed with PEA/MBA spacer cations. In these chiral perovskites, a group of four interconnected PbI_6 octahedra forms a puckered square pattern due to the tilting of adjacent octahedra. This tilting causes the equatorial Pb-I-Pb bond angles to deviate notably from the ideal 180° characteristic of an undistorted perovskite sheet. All chiral perovskites exhibit tilting asymmetry, resulting in two distinct equatorial Pb-I-Pb bond angles of 143.6° and 151.8° (Supplementary Fig. 19), which correspond to two different equatorial iodine atoms. The disparity in these equatorial bond angles within chiral perovskite quantum wells is likely a consequence of asymmetric hydrogen bonding involving the in-plane iodine atoms of the $[\text{PbI}_4]^{2-}$ layers.

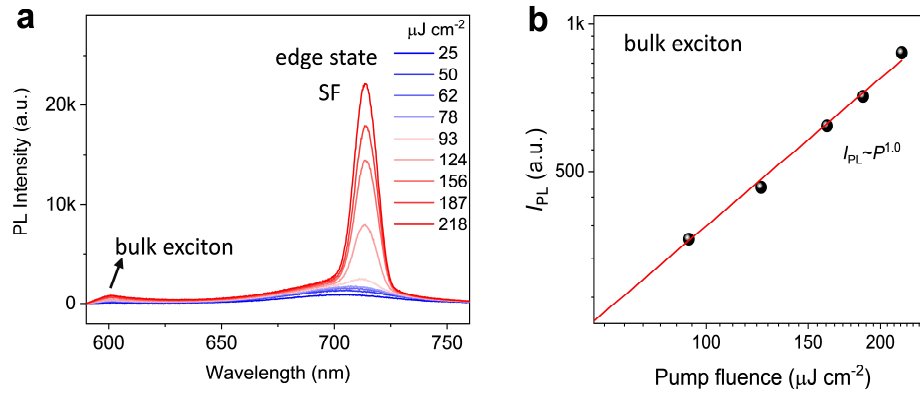
The equatorial iodine atoms undergo out-of-plane distortions that spiral around the 2_1 screw axis, which aligns with the b-axis (Supplementary Fig. 19), as the chiral perovskite quantum well crystallizes into a chiral space group. Between the two enantiomers, the spirals formed by the bonds between the two types of iodine, mediated by adjacent Pb atoms, display opposite yet distinct helicities. Consequently, the net crystallographic helicity of the iodine distortions in SMBA is opposite to that in RMBA, as previously discussed.³³



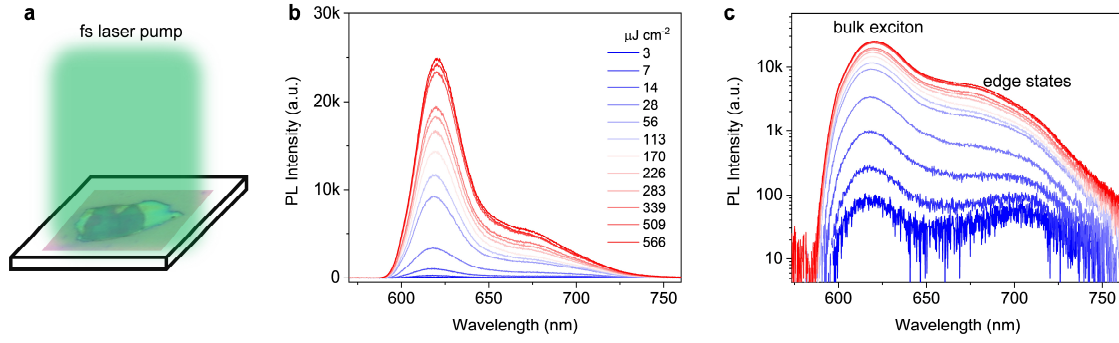
Supplementary Fig. 1 | Close-up scanning electron microscopy images. Scanning electron microscope (SEM) images of the **a-b**, SMBA, **c-d**, RMBA and **e-f**, PEA perovskite superlattices.



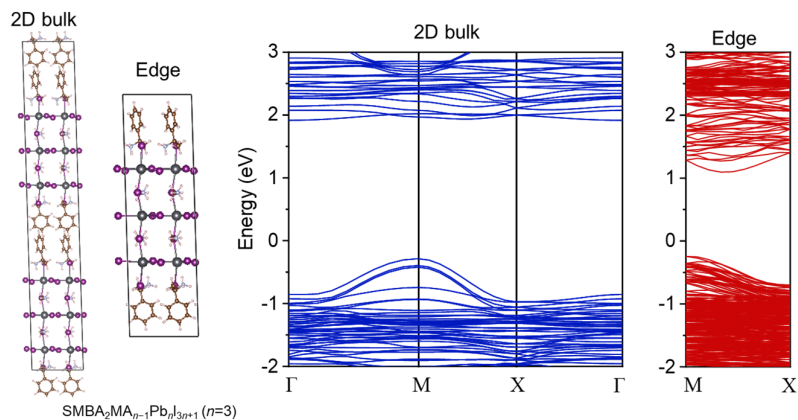
Supplementary Fig. 2 | Wide angle scanning electron microscopy images. SEM images of **a**, SMBA, **b**, RMBA and **c**, PEA perovskite superlattices.



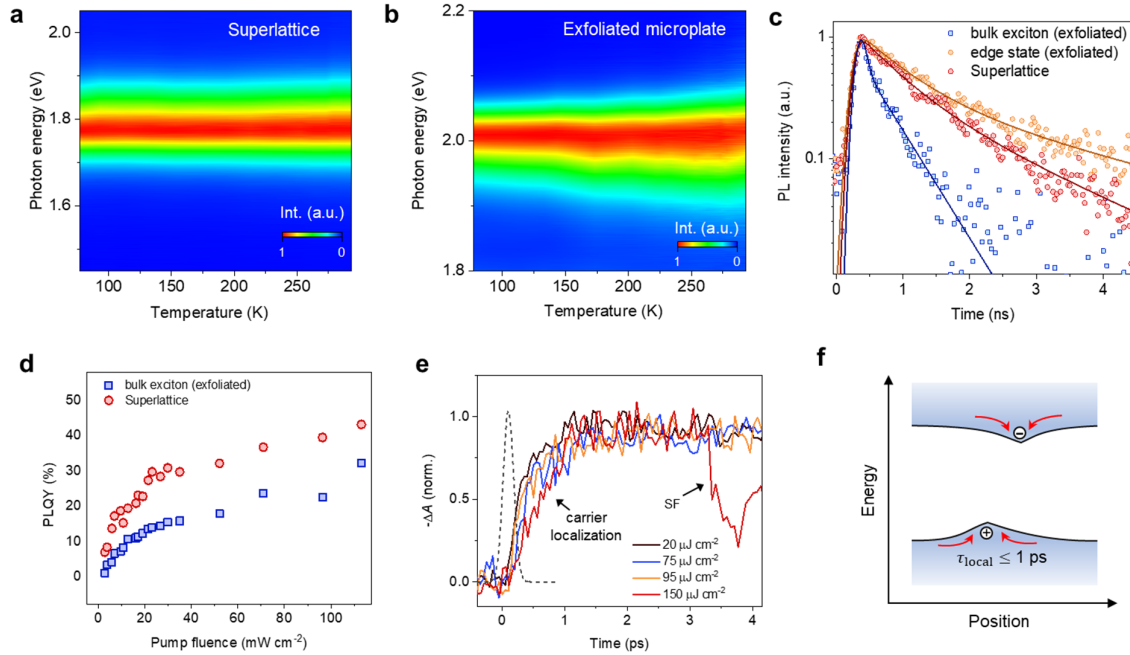
Supplementary Fig. 3 | Weak bulk exciton emission from superlattices. a. Pump fluence dependent PL spectra of SMBA superlattices with a broad wavelength collection range. **b.** Bulk exciton emission at 600 nm exhibits a linear dependence on pump fluence.



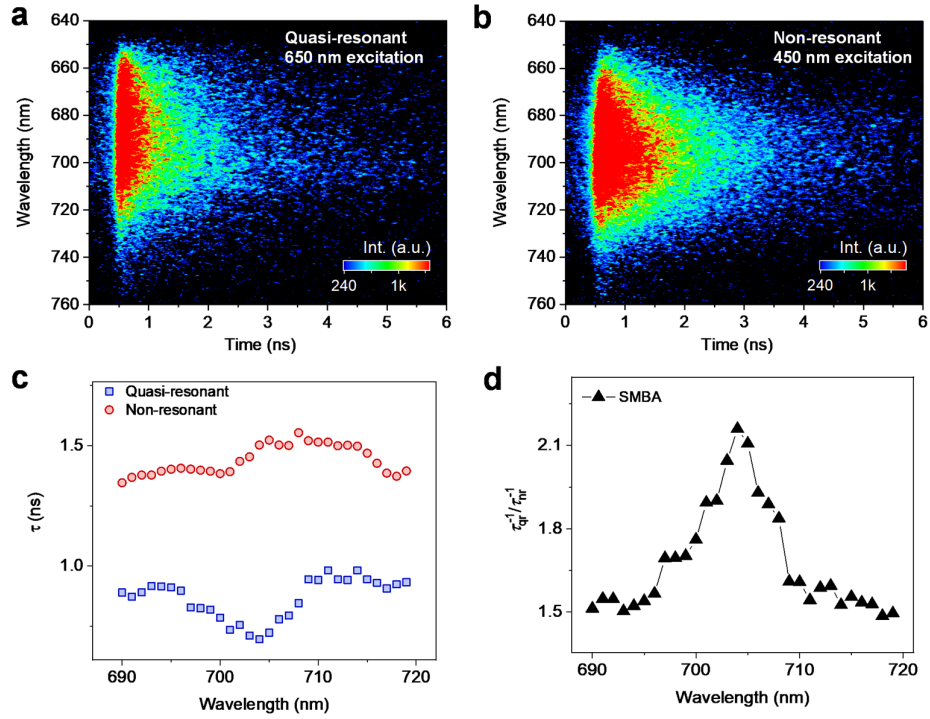
Supplementary Fig. 4 | Power-dependent PL of an exfoliated chiral perovskite single-crystal microplate. **a**, Schematic of the measurement configuration, where the laser beam illuminates the entire flake including its edges. Power-dependent PL spectra of the $(\text{PEA})_{1.2}(\text{SMBA})_{0.8}(\text{MA})_2\text{Pb}_3\text{I}_{10}$ single-crystal flake plotted on linear (**b**) and logarithmic (**c**) scales. Neither amplified spontaneous emission (ASE) nor SF is observed from either the bulk exciton or the edge state emission.



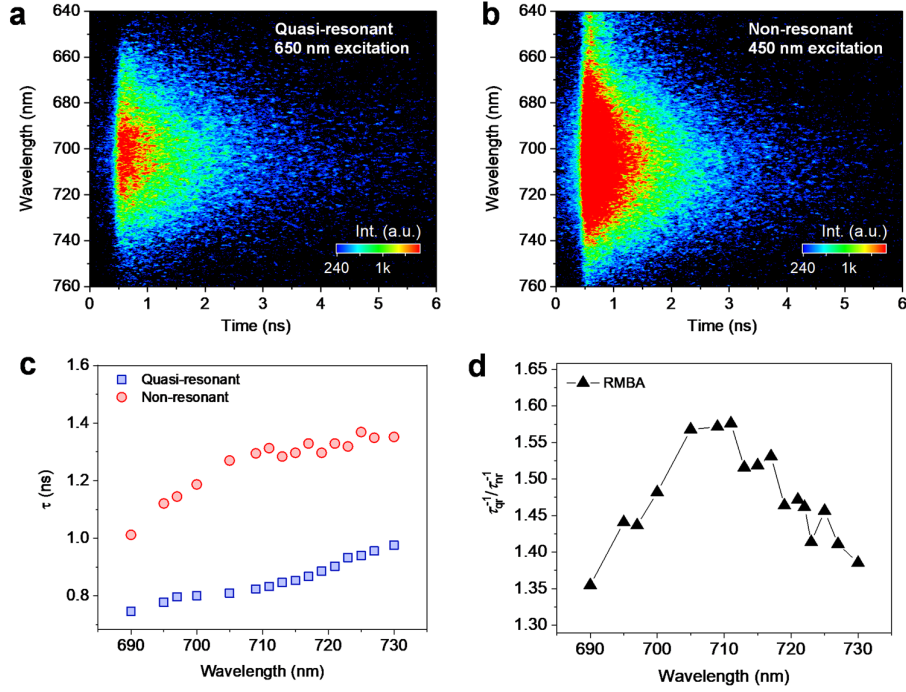
Supplementary Fig. 5 | Density functional theory (DFT) calculated band structures of 2D bulk and edges of pure chiral perovskites ($\text{SMBA}_2\text{MA}_{n-1}\text{Pb}_n\text{I}_{3n+1}$ ($n = 3$)) with optimized slab models for simulation. The calculated bandgap of the 2D bulk is 2.20 eV, whereas the edge state bandgap is 1.34 eV.



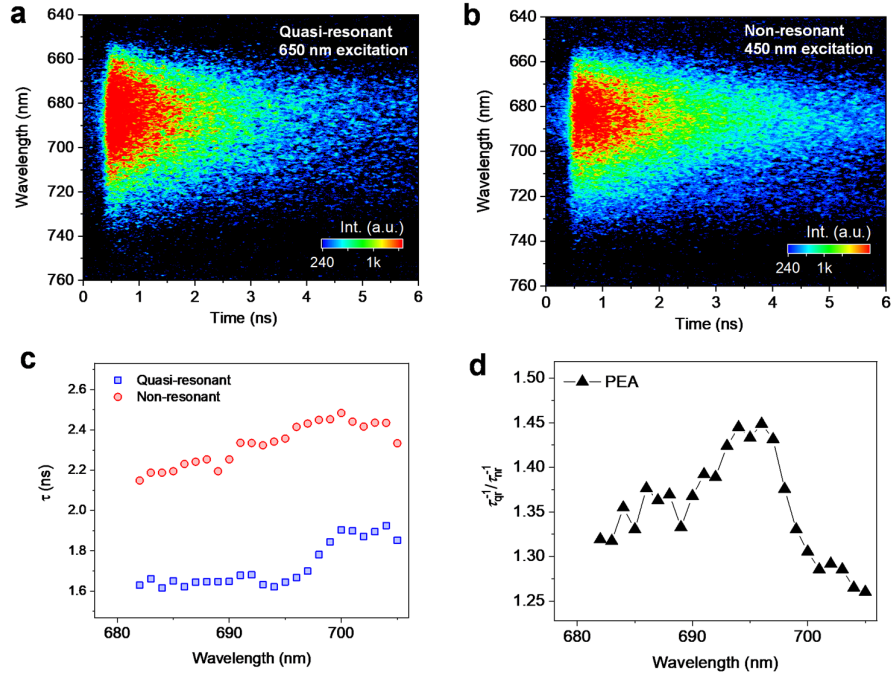
Supplementary Fig. 6 | Comparison between perovskite superlattices and exfoliated microplates. 2D plot of the normalized PL spectra of **a**, SMBA perovskite superlattices (edge state emission) and **b**, exfoliated SMBA perovskite microplate ($n = 3$) measured at the center (bulk exciton emission). **c**, Time resolved PL traces of bulk exciton emission at 620 nm at the center, and edge state emission at 700 nm from the exfoliated microplate. The emission of SMBA perovskite superlattices at 700 nm is plotted for comparison. **d**, PLQY verses pump fluence of bulk exciton emission at 620 nm and SMBA perovskite superlattice emission at 700 nm (pump laser: 537 nm CW laser). **e**, Normalized TA traces demonstrating the initial population buildup probed at the SF position (705 nm) of the SMBA perovskite superlattices. The dashed curve represents the instrument response, which exhibits much faster rise dynamics, confirming that the observed slow rise is intrinsic and not an artifact of the system. **f**, Schematic of photoexcited carrier localization at the edge states of quasi-2D perovskites.



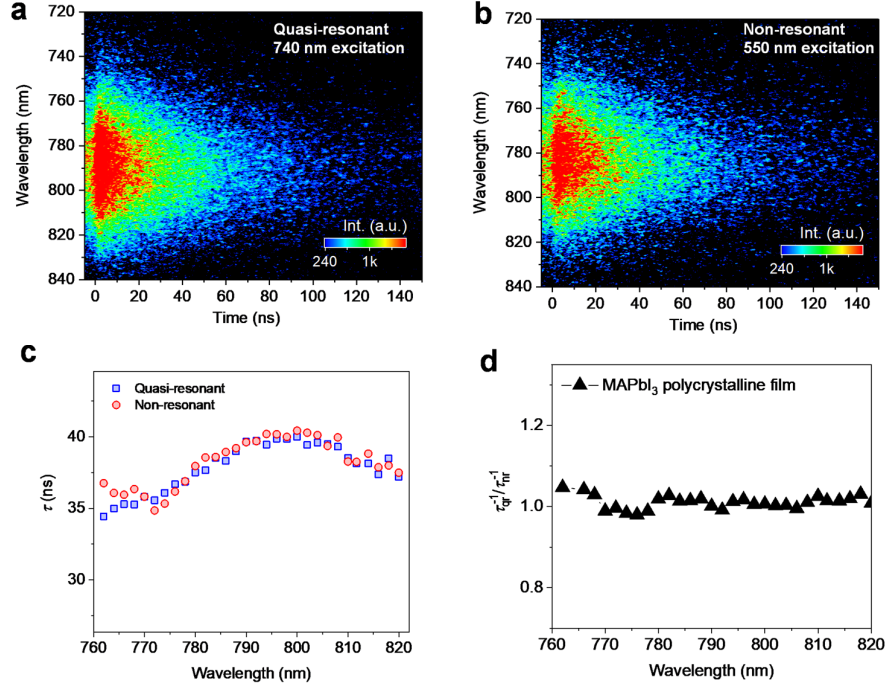
Supplementary Fig. 7 | Excitation energy dependent emission recombination rate (SMBA perovskite superlattices). Streak camera images under **a**, 650 nm quasi-resonant excitation and **b**, 420-nm non-resonant excitation ($\sim 5 \mu\text{J cm}^{-2}$) pumped below the SF threshold. **c**, PL decay time (lifetime at $1/e$ of peak intensity) as a function of wavelength under quasi-resonant and non-resonant excitation. **d**, Spectral dependence of the ratio $\tau_{qr}^{-1}/\tau_{nr}^{-1}$ between quasi-resonant and non-resonant radiative decay rates.



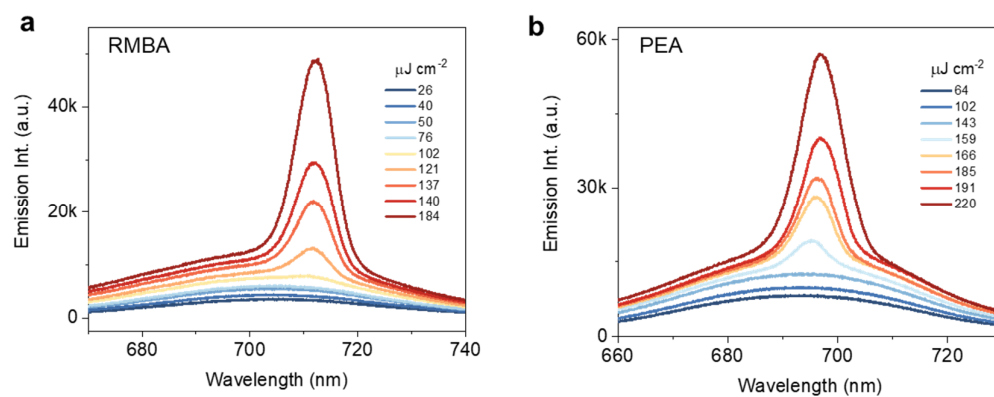
Supplementary Fig. 8 | Excitation energy dependent emission recombination rate (RMBA perovskite superlattices). Streak camera images under **a**, 650 nm quasi-resonant excitation and **b**, 420 nm non-resonant excitation ($\sim 5 \mu\text{J cm}^{-2}$) pumped below the SF threshold. **c**, PL decay time (lifetime at $1/e$ of peak intensity) as a function of wavelength under quasi-resonant and non-resonant excitation. **d**, Spectral dependence of the ratio $\tau_{qr}^{-1}/\tau_{nr}^{-1}$ between quasi-resonant and non-resonant radiative decay rates.



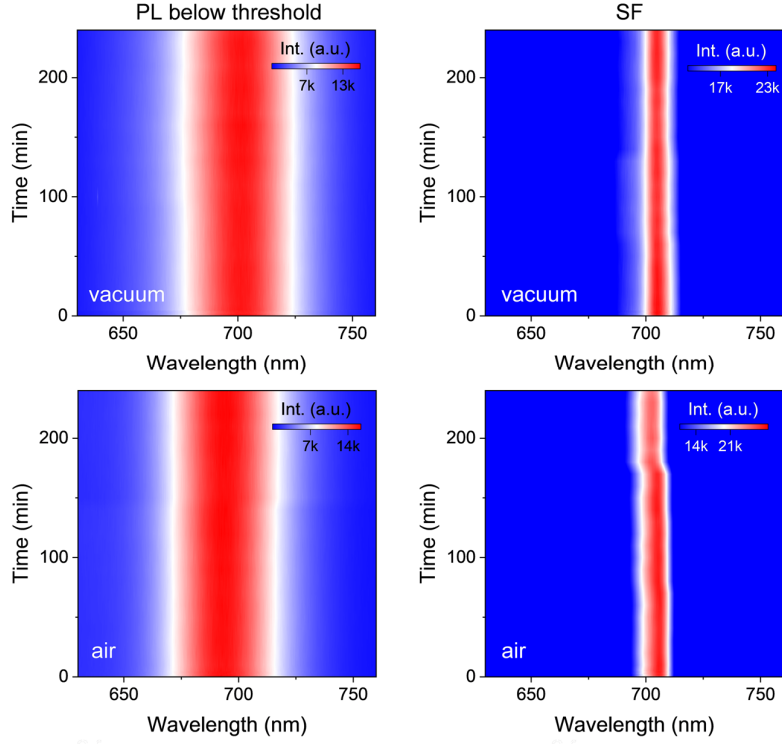
Supplementary Fig. 9 | Excitation-energy dependent emission recombination rate (PEA perovskite superlattices). Streak camera images under **a**, 650 nm quasi-resonant excitation and **b**, 420 nm non-resonant excitation ($\sim 5 \mu\text{J cm}^{-2}$) pumped below the SF threshold. **c**, PL decay time (lifetime at $1/e$ of peak intensity) as a function of wavelength under quasi-resonant and non-resonant excitation. **d**, Spectral dependence of the ratio $\tau_{qr}^{-1}/\tau_{nr}^{-1}$ between quasi-resonant and non-resonant radiative decay rates.



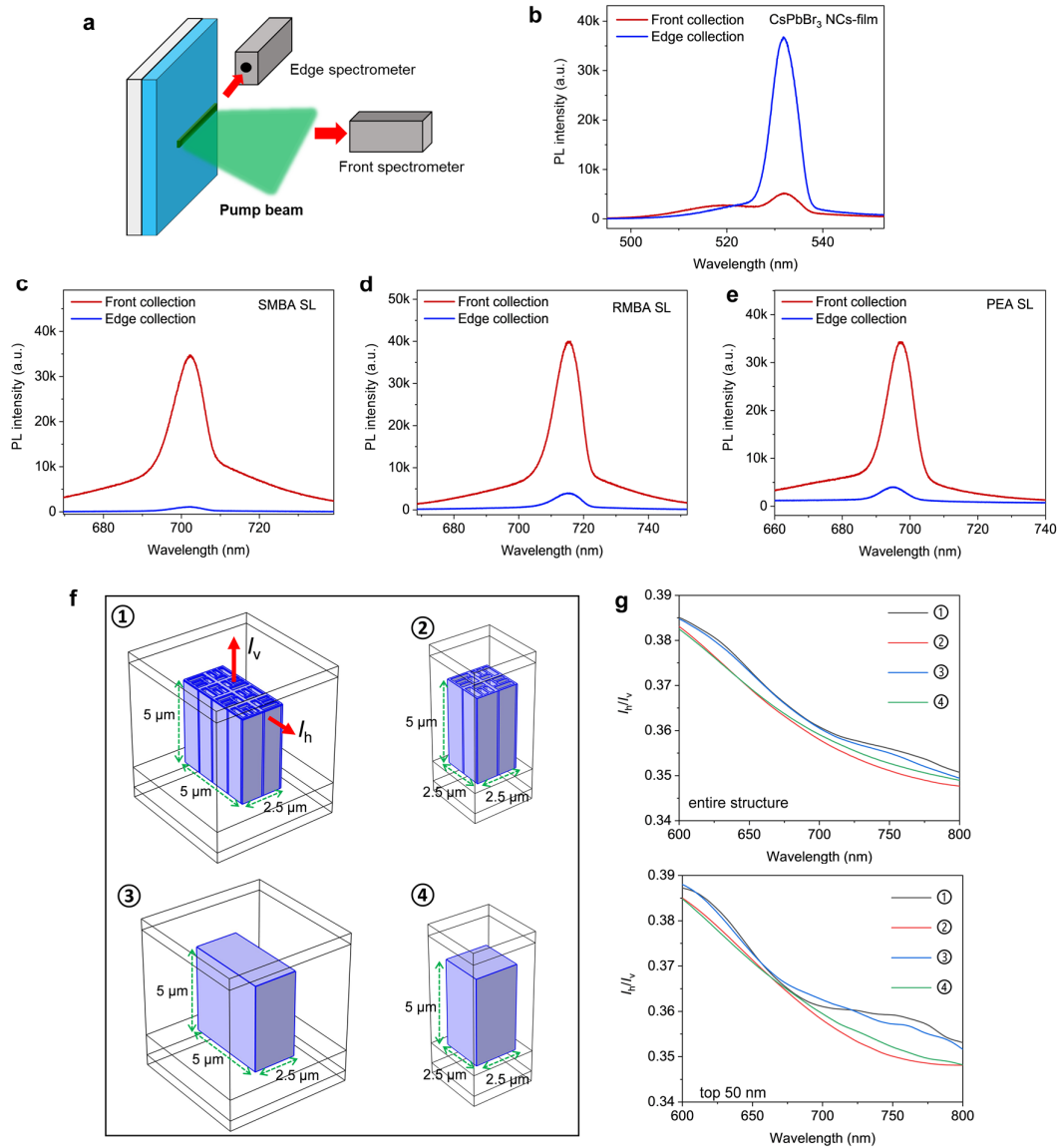
Supplementary Fig. 10 | Excitation-energy dependent emission recombination rate (MAPbI₃ polycrystalline film on glass substrate). Streak camera images under **a**, 740 nm quasi-resonant excitation and **b**, 550 nm non-resonant excitation ($5 \mu\text{J cm}^{-2}$). **c**, PL decay time as a function of wavelength under quasi-resonant and non-resonant excitation. **d**, Spectral dependence of the ratio $\tau_{qr}^{-1}/\tau_{nr}^{-1}$ between quasi-resonant and non-resonant radiative decay rates.



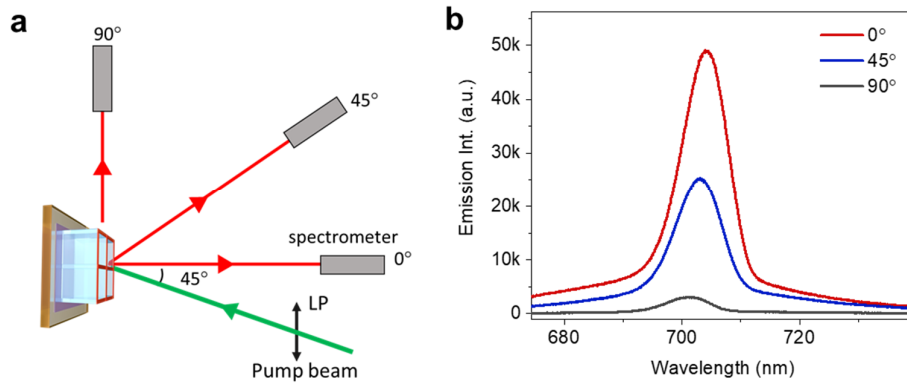
Supplementary Fig. 11 | Emission spectra of the RMBA and PEA perovskite superlattices. Pump fluence dependent emission spectra for the **a**, RMBA and **b**, PEA perovskites. Colors denote different pump fluences.



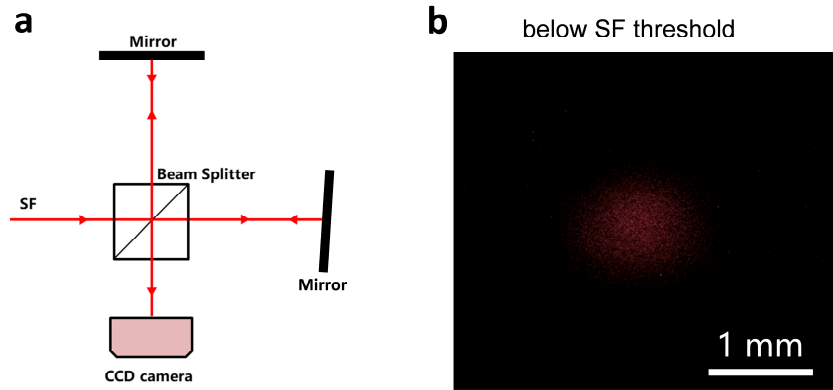
Supplementary Fig. 12 | Stability measurements. 2D color plot of PL spectra of SMBA perovskite superlattices as a function of time under 550 nm 1 kHz fs laser excitation. Measurements were performed in vacuum (10^{-3} Pa; top panels) and in ambient air (relative humidity $\sim 50\%$; bottom panels), with pump fluences below the SF threshold ($53 \mu\text{J cm}^{-2}$; left panels) and above the SF threshold ($98 \mu\text{J cm}^{-2}$; right panels).



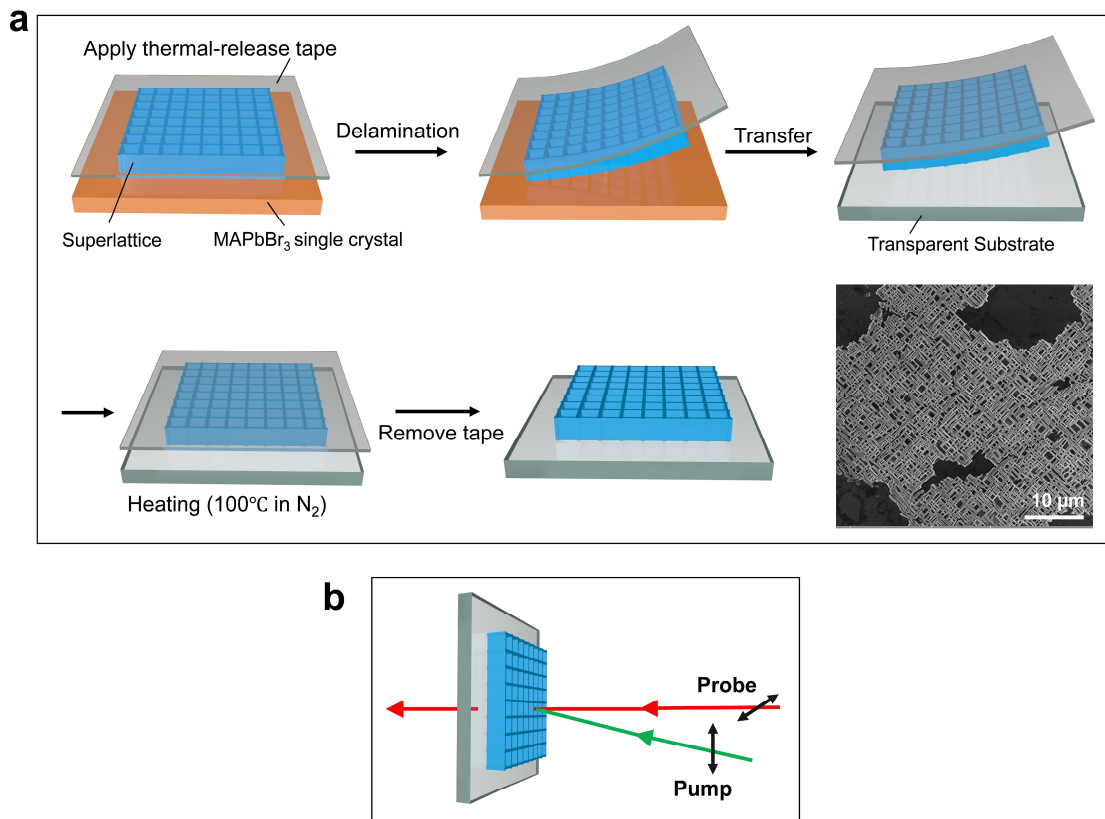
Supplementary Fig. 13 | Directional SF under stripe-shaped beam excitation. **A**, Schematic experimental setup for measuring SF or ASE at the sample edge and in the forward direction under a stripe-shaped beam excitation. The beam was focused by a cylindrical lens with dimension of 0.2 mm × 5 mm. **b**, Control ASE spectra of the CsPbBr₃ nanocrystal (NCs) film. SF spectra of **c**, SMBA, **d**, RMBA, and **e**, PEA perovskite superlattices. **f**, Schematic of the perovskite structures used in numerical simulations comparing the crisscross superlattices to bulk crystals. The dimensions of each structure are indicated. **g**, Ratio of far-field emission intensity in the horizontal direction to that in the vertical direction (I_h/I_v) for the different perovskite structures, calculated by treating either the entire structure (top) or only the top 50 nm (bottom) as the emitting region.



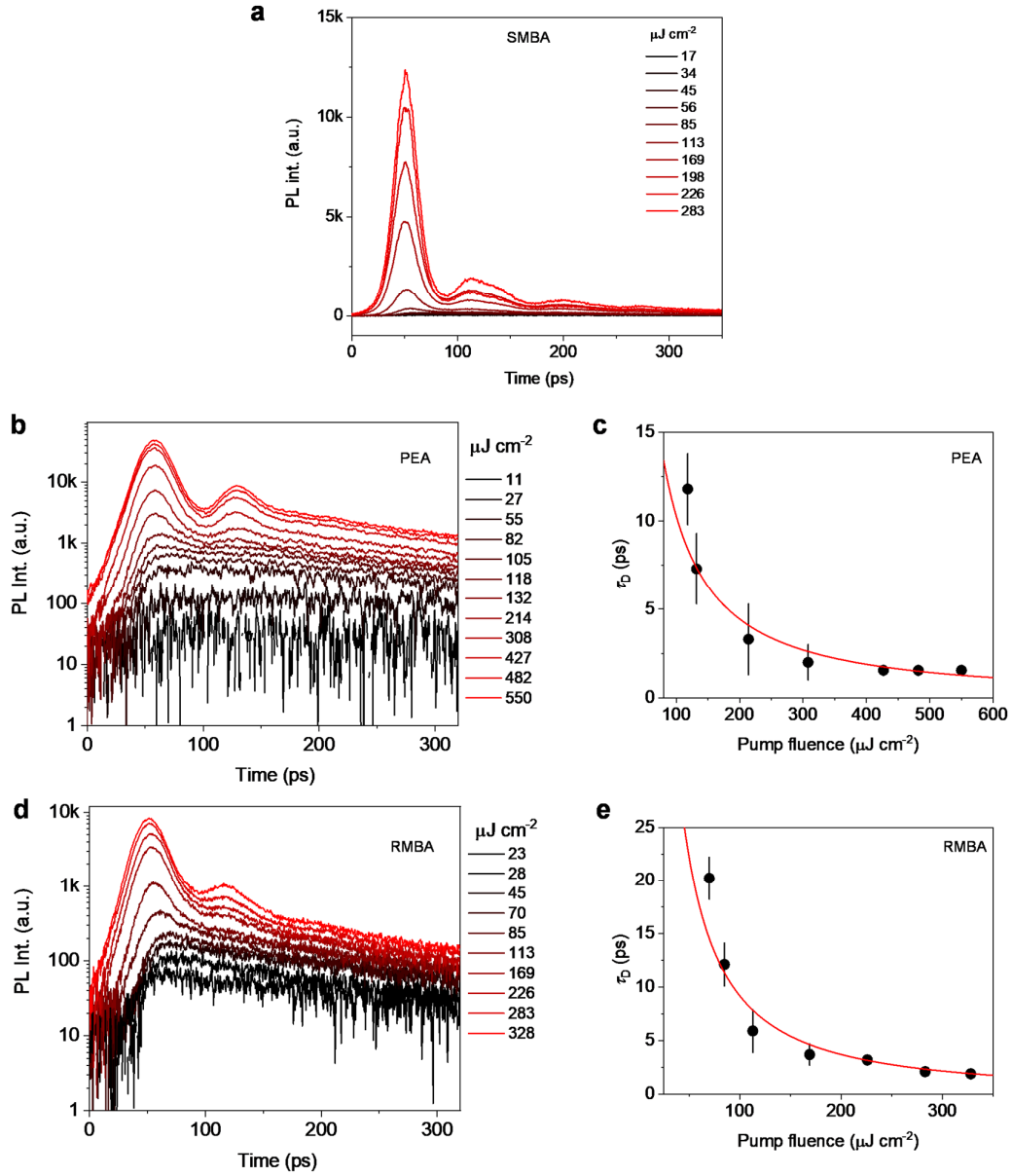
Supplementary Fig. 14 | Directional SF under point excitation. **a**, Schematic experimental setup for SF emission collected at 0, 45, and 90 degrees relative to the surface normal of SMBA perovskite superlattices on the MAPbBr₃ single crystal substrate. **b**, Collected SF spectra under 550 nm excitation.



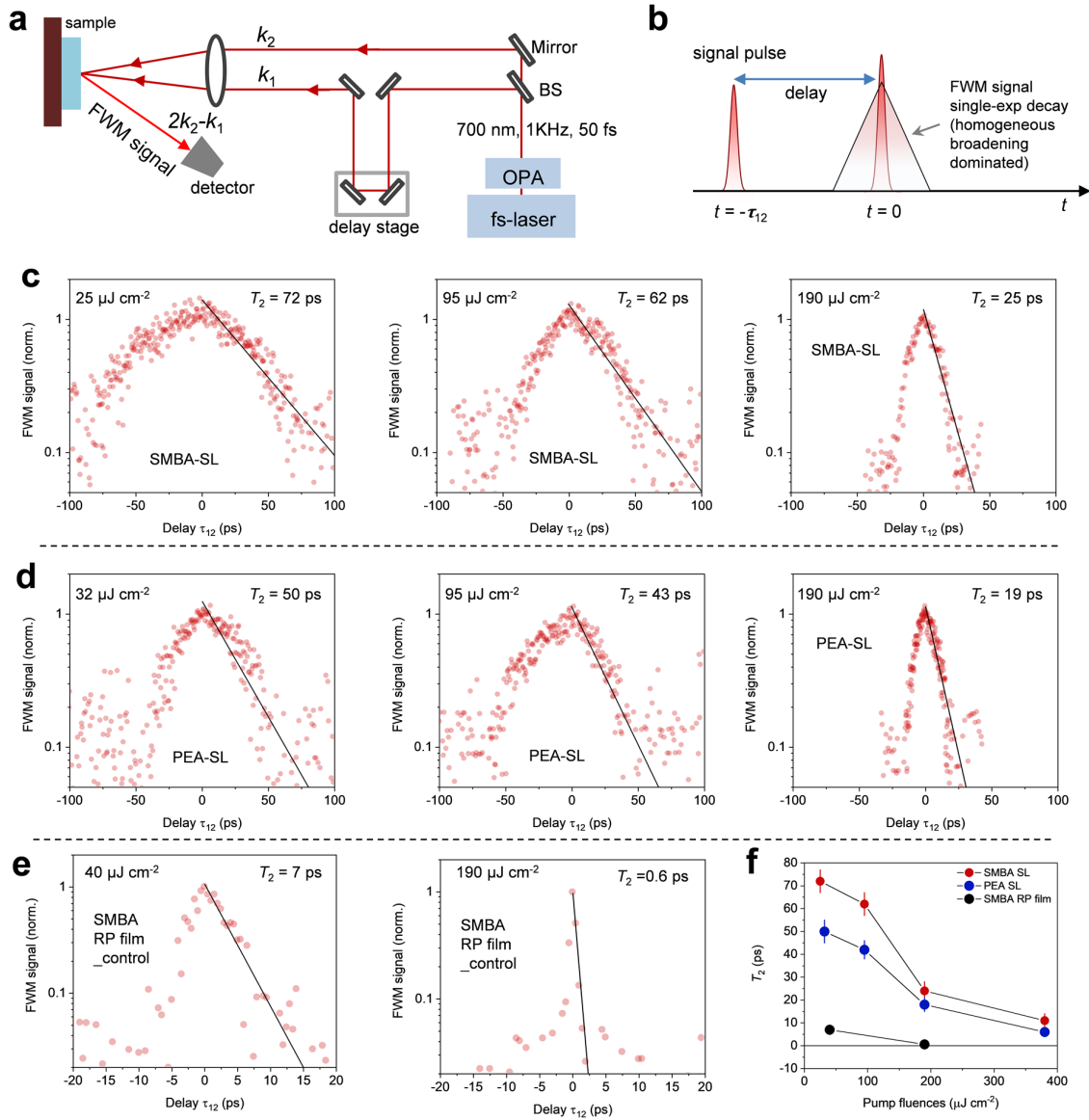
Supplementary Fig. 15 | Coherence measurements recorded with a Michelson interferometer. **a**, Schematic Michelson interferometer setup. One of the mirrors is tilted slightly to produce spatial coherence fringes. **b**, Emission interferograms recorded below the SF threshold do not show any interference fringes.



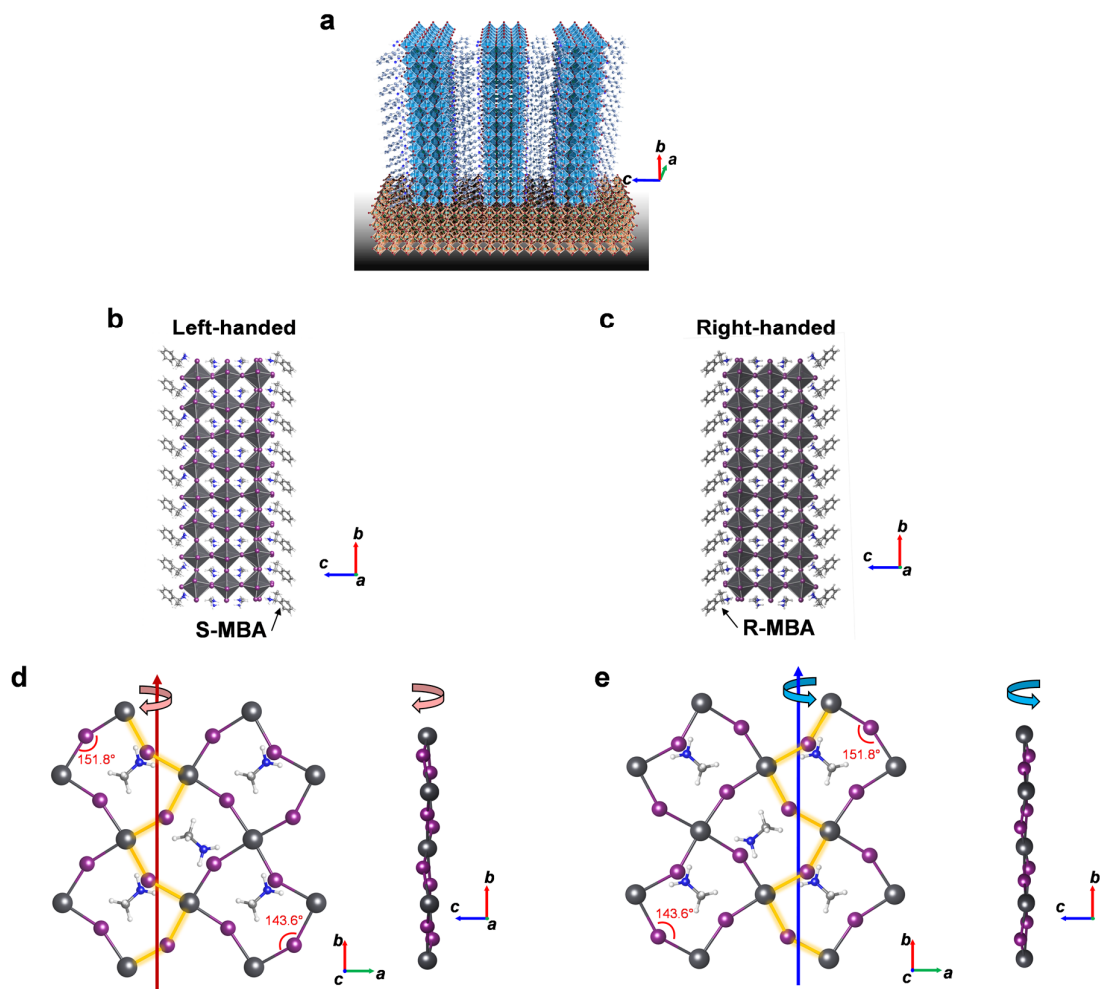
Supplementary Fig. 16 | Superlattice transfer process and pump-probe measurement setup. a, Schematic illustration of the steps for transferring perovskite superlattices grown on a MAPbBr₃ single crystal to a transparent quartz substrate. A scanning electron microscope (SEM) image of the transferred superlattices is included. **b**, Schematic of the pump-probe transient absorption measurement setup with cross-polarization, configured in a transmission geometry.



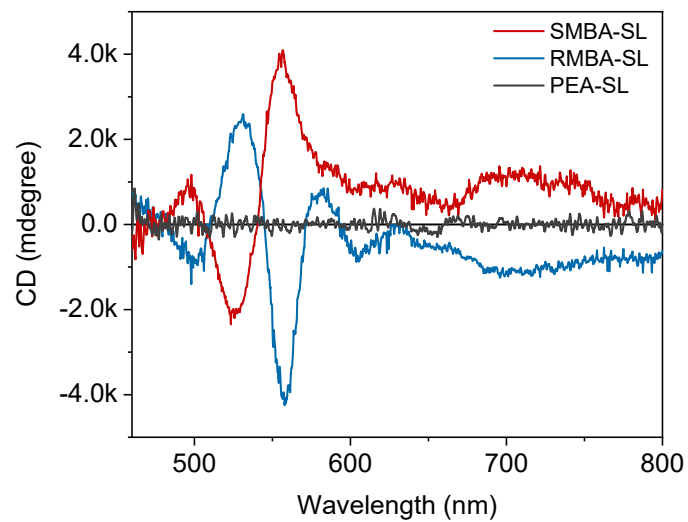
Supplementary Fig. 17 | Power-dependent time-resolved photoluminescence and delay time analysis. **a**, Linear-scale plot of time-resolved PL spectra (original data in Fig. 2b in main text) for SMBA perovskite superlattices as a function of pump fluence. **(b, d)** Log-scale plots of time-resolved PL spectra (original data in Extended Data Figs. 6 and 7) for **(b)** PEA and **(d)** RMBA perovskite superlattices as a function of pump fluence. **(c, e)** Extracted SF delay times (τ_D) as a function of pump fluence for **(c)** PEA and **(e)** RMBA superlattices, respectively. Solid curves represent fits to the relation $\tau_D \propto 1/\Gamma \ln(N)/(N + 1)$, where $N = bP^\alpha$, with $\alpha = 1.49$ for PEA and $\alpha = 1.51$ for RMBA.



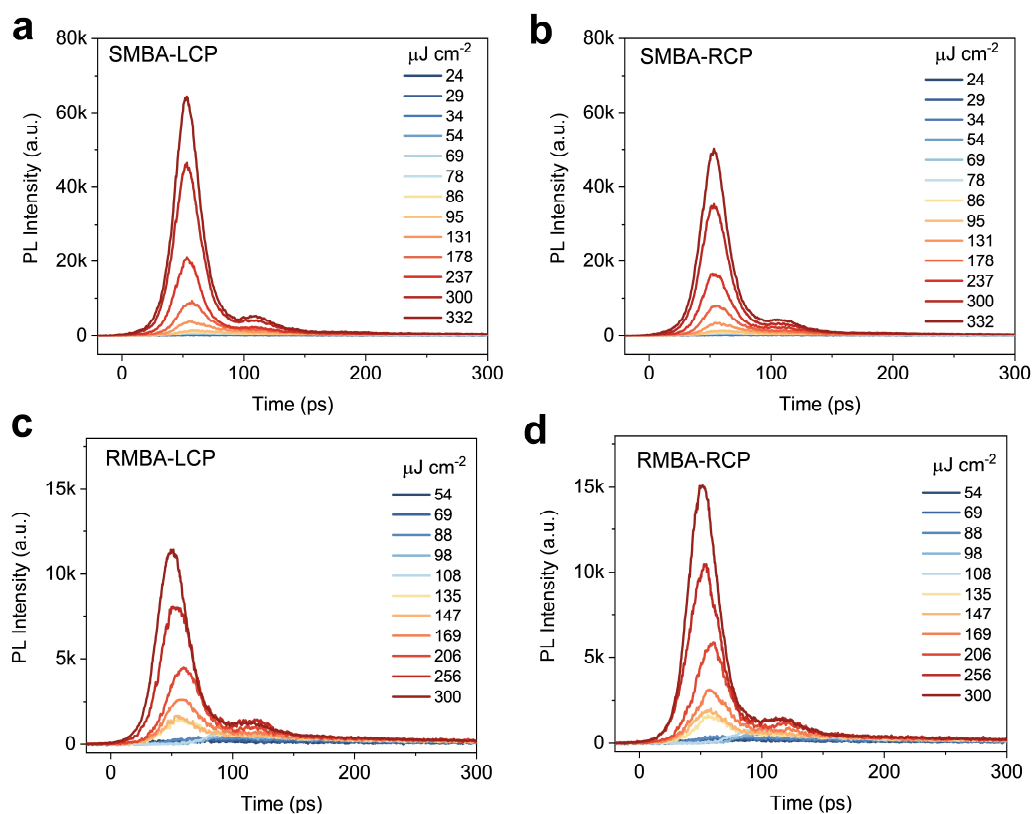
Supplementary Fig. 18 | T_2 coherence time measurements. **a**, Schematic of the transient four-wave mixing (FWM) experimental setup. **b**, Illustration of the FWM signal generation principle. Transient FWM signals from **(c)** SMBA superlattice (SMBA-SL), **(d)** PEA superlattice (PEA-SL), and **(e)** a control SMBA Ruddlesden–Popper (RP) perovskite thin film on quartz substrate. Solid lines in **c–e** are single-exponential decay fits. **f**, Extracted T_2 dephasing time as a function of pump fluence, measured under 1 kHz repetition rate, 50 fs pulse duration, and 700 nm excitation. Error bars represent the fitting uncertainty.



Supplementary Fig. 19 | Chirality-transfer from the chiral ligand to the perovskite superlattices. **a**, Schematic vertical chiral perovskite superlattice grown on MAPbBr₃ single crystal substrate. Schematic cross-sectional view of **(b)** SMBA and **(c)** RMBA chiral perovskites (S/R-MBA₂MA₂Pb₃I₁₀) ($n = 3$) structures created by VESTA software³⁴. Top [PbI₄]²⁻ layers in **(d)** SMBA and **(e)** RMBA chiral perovskites. The out-of-plane helical distortions (indicated by curved arrows) can be seen from the spiraling of corresponding Pb–I–Pb bonds (marked by thicker orange lines) around the *b*-screw axes (red and blue arrows). The gray and purple spheres denote Pb, I atoms, respectively. Note: For clarity in illustrating chirality transfer, only the structures with pure chiral ligands are shown, although the actual superlattices contain a mixture of chiral (MBA) and achiral (PEA) ligands (see Methods).

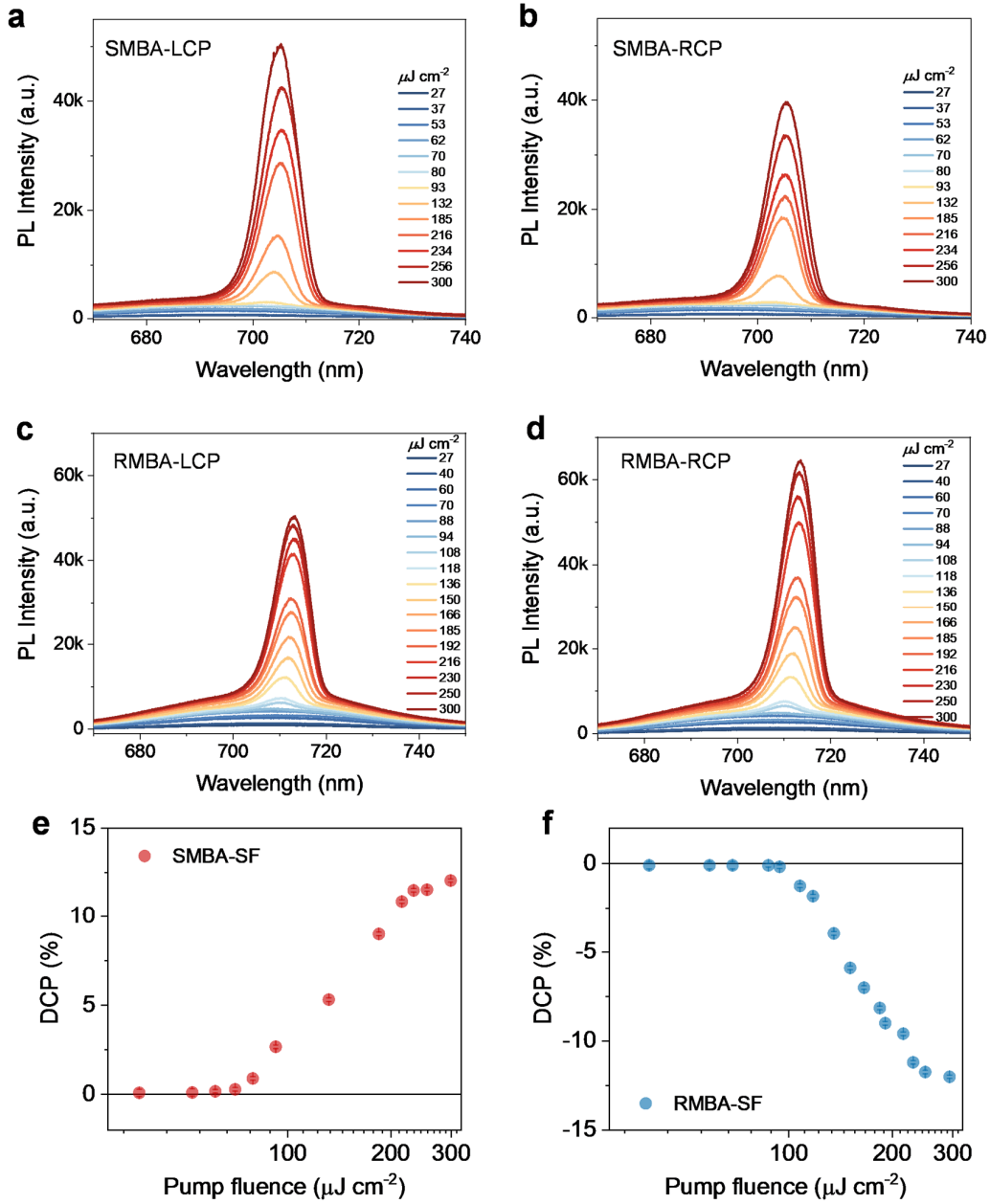


Supplementary Fig. 20 | Circular dichroism spectra of achiral PEA and chiral SMBA and RMBA perovskite superlattices (SL).

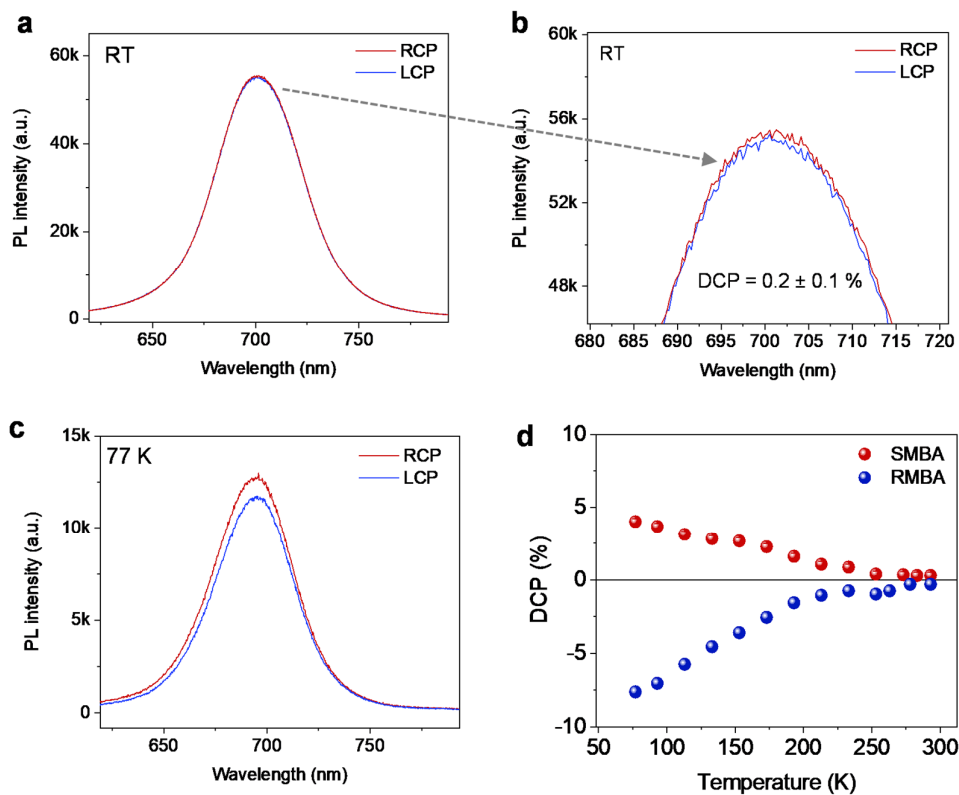


Supplementary Fig. 21 | Time-resolved PL spectra of chiral perovskite superlattices.

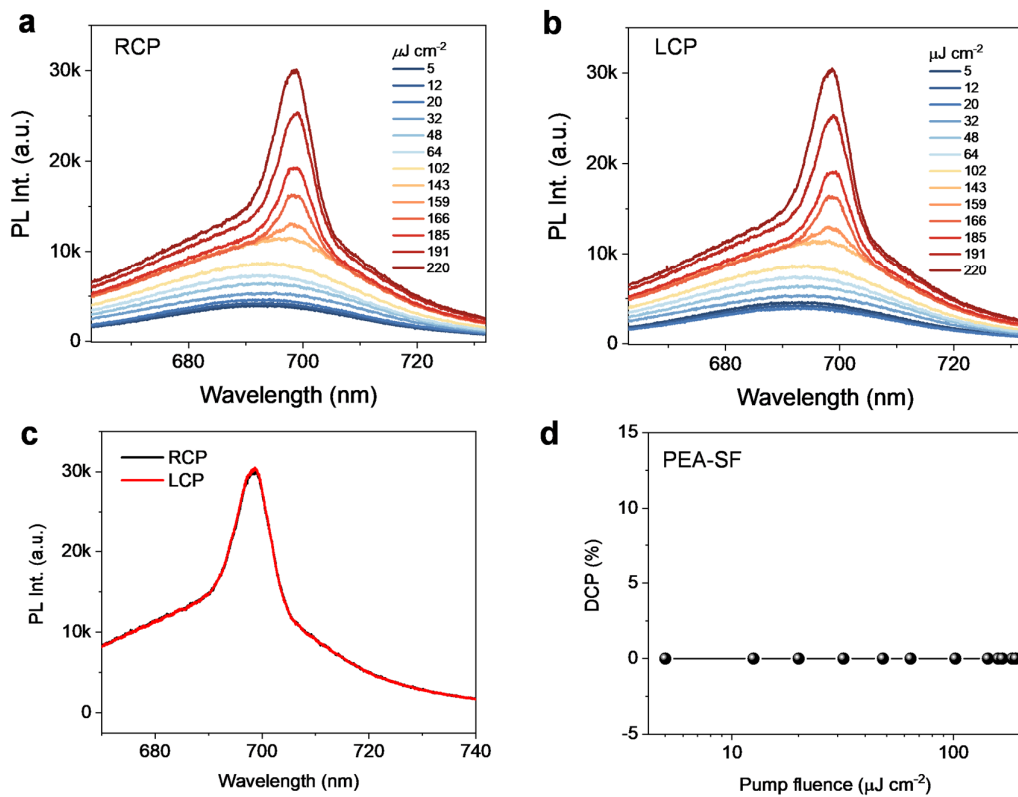
a,b, Left-circularly polarized (LCP, **a**) and right-circularly polarized (RCP, **b**) TRPL spectra of the SMBA superlattices. **c,d**, LCP (**c**) and RCP (**d**) PL spectra of the RMBA superlattices under linearly polarized excitation at 550 nm.



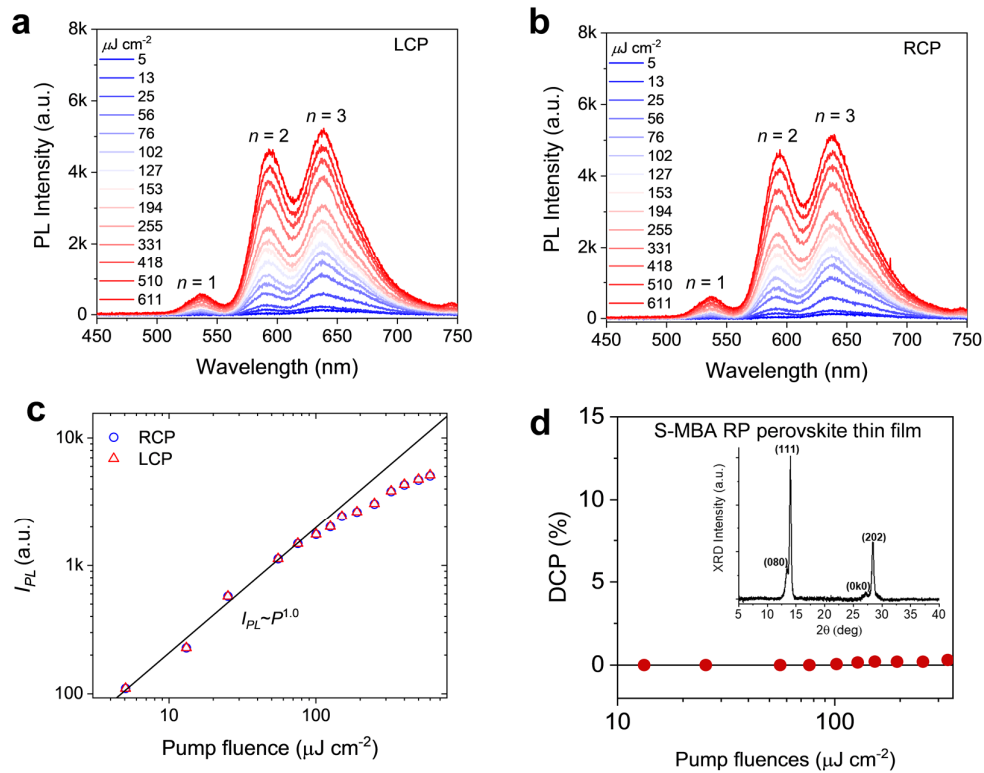
Supplementary Fig. 22 | Time-integrated PL spectra and degree of circular polarization (DCP) of chiral perovskite superlattices. Left-circularly polarized (LCP, **a**) and right-circularly polarized (RCP, **b**) PL spectra of the SMBA superlattices. LCP (**c**) and RCP (**d**) PL spectra of the RMBA superlattices under linearly polarized excitation at 550 nm. DCP of the SF of (**e**) SMBA and (**f**) RMBA superlattices as a function of pump fluence. The slightly higher values of DCP_0 compared to the time-integrated DCP are attributed to a reduction in the polarization degree at times beyond the SF peak.



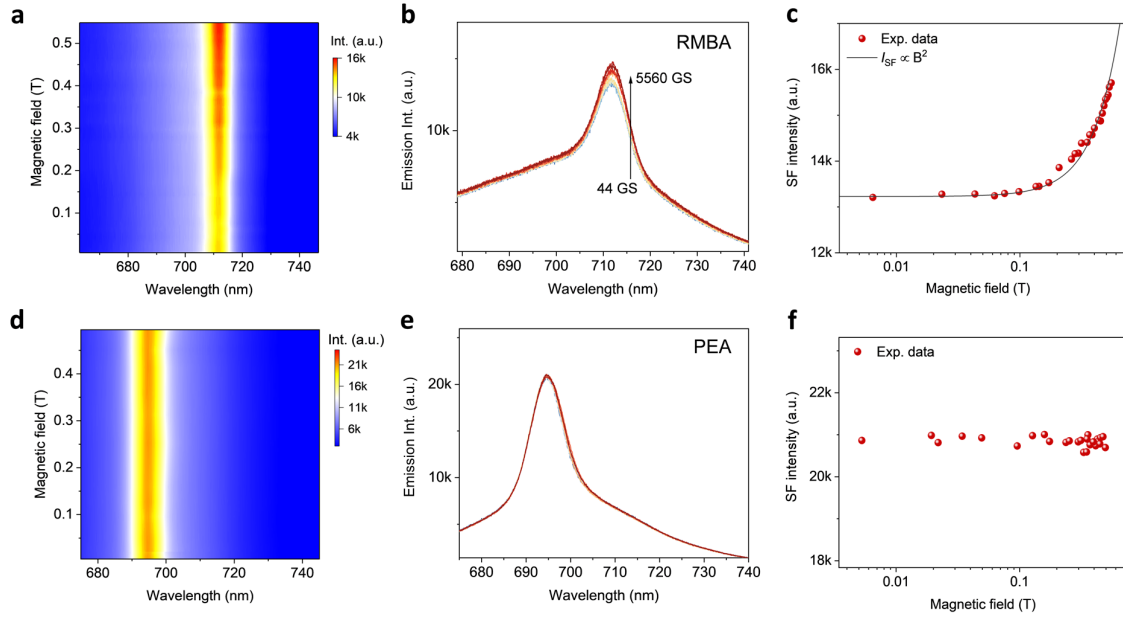
Supplementary Fig. 23 | Circularly polarized PL spectra at (a, b) room temperature and (c) 77K of RMBA perovskite superlattices. d, DCP of spontaneous emission of S/RMBA perovskite superlattices as a function of temperature (at $40 \mu\text{J cm}^{-2}$).



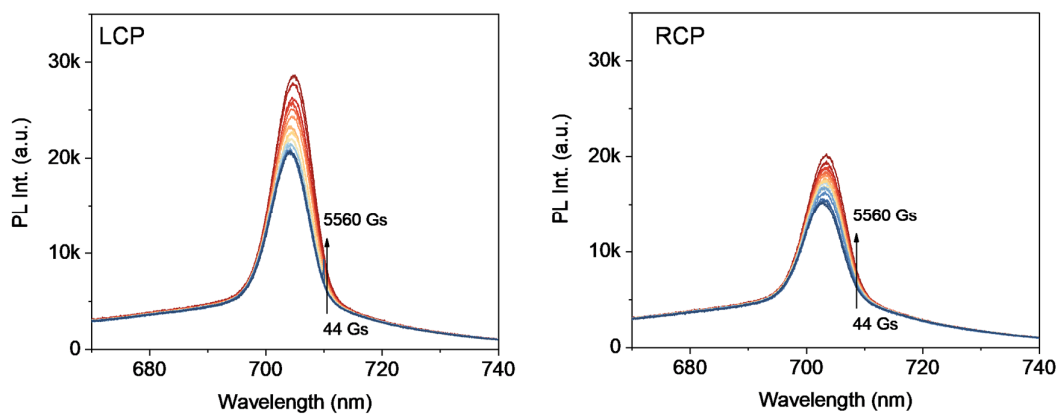
Supplementary Fig. 24 | Absence of chiral SF in achiral PEA perovskite superlattices. PL spectra from an achiral PEA perovskite superlattice under 550 nm linearly polarized excitation, monitored in **(a)** RCP and **(b)** LCP detection. **(c)** Comparison of SF spectra at a pump fluence of 220 $\mu\text{J cm}^{-2}$. **(d)** DCP plotted as a function of pump fluence.



Supplementary Fig. 25 | Power dependent PL spectra of SMBA Ruddelsden-Popper (RP) perovskite thin film control sample. Time-integrated PL spectra for left-circularly polarized (LCP, **a**) and right-circularly polarized (RCP, **b**) emission. The peaks correspond to $n = 1, 2, 3$ phases, respectively³⁵, and no SF is observed. **c**, Peak intensity at 650 nm versus pump fluence, showing a linear dependence. **d**, Degree of circular polarization (DCP) versus pump fluence. Neither SF nor DCP amplification is observed in the chiral control sample. Inset of **d**, XRD pattern of the thin film, revealing the RP perovskite phases.



Supplementary Fig. 26 | Magnetic-field tunable SF from RMBA and PEA perovskite superlattices. 2D contour color plots of SF spectra at various external magnetic fields for (a-b) RMBA and (d-e) PEA perovskite superlattices, respectively, pumped above the SF threshold ($150 \mu\text{J cm}^{-2}$ for RMBA, $195 \mu\text{J cm}^{-2}$ for PEA). Emission strength versus magnetic field strength B for (c) RMBA and (f) PEA perovskite superlattices, respectively.



Supplementary Fig. 27 | Magnetic field tunable SF in the SMBA perovskite superlattice.
a, left- and **b**, right- circularly polarized SF spectra of SMBA perovskite superlattices under an external magnetic field at a pump fluence above the SF threshold.

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