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Peer Review File

Chiral superfluorescence from perovskite superlattices at room temperature

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This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Referee #1

(Remarks to the Author)
The authors report the realization of strong left- and right-handed circularly polarized superfluorescence (CP-SF) from large-area (>100 μm × 100 μm) vertically aligned chiral perovskite quantum well superlattices at room temperature. Supported by theoretical quantum optics calculations, they claim that chirality-induced photon transport facilitates the transition to CP-SF from initially incoherent, unpolarized spontaneous emission, resulting in an amplified degree of circular polarization of up to approximately 14%. The application of a low external magnetic field can further modulate the degree of CP-SF.
Achieving high-temperature cooperative emission is an extremely attractive goal, with the potential to unlock quantum photonic applications by reducing system complexity and eliminating the need for cryogenic apparatus. In addition, the claimed observation of circularly polarized SF could stimulate further research in the fields of quantum optics and spintronics. However, the main claim, the observation of SF at room temperature, presents a fundamental challenge: how to retain the coherence properties of excitons in solid-state compounds can be retained and protected against decoherence processes at elevated temperatures. Despite its promise, the current manuscript fails to provide robust evidence to substantiate the very main claim.
Here are more detailed comments:
1. There is no direct experimental proof of exciton coherence, which is crucial when invoking coherent coupling.
2. Oscillations in the time domain are not exclusively indicative of superfluorescence (SF). They can also arise from peculiar intraband relaxation in stimulated emission experiments, as discussed in J. Phys. B: At. Mol. Opt. Phys., 39, 215 (2006), and J. Appl. Phys., 91, 6367 (2002). A more detailed analysis is needed to rule out the amplified spontaneous emission (ASE) process.
3. The claimed 14% degree of circular polarization is significantly lower than the values recently reported for chiral ASE and lasing (see for example, https://doi.org/10.1002/adma.202301573, https://pubs.acs.org/doi/full/10.1021/acs.nanolett.4c03838). What could be the benefit of CP-SF?
4. The reported 1 mm-long spatial coherence seems incompatible with a single SF domain. This is more likely to originate from a lasing/photonic mode.
5. The transient peak intensity at 705 nm increases superlinearly with pump fluence, with an exponent greater than 2. This behavior is inconsistent with SF buildup and is more likely associated with stimulated emission. Although the authors invoke bimolecular recombination (which is less likely in these compounds due to strong quantum confinement and large binding energies), a detailed analysis to support this claim is missing.
6. The change in decay time as a function of excitation density is rather abrupt, as expected for the transition from spontaneous to stimulated emission. Please note that the 120 ps decay time at room temperature is likely largely dominated by non-radiative processes.
7. It is unclear why spontaneous emission is not circularly polarized, as reported for similar compounds (https://www.nature.com/articles/s41467-020-18485-7, https://pubs.acs.org/doi/10.1021/acsnano.9b00302). As claimed by the authors, there should be net crystallographic helicity due to the chiral ligand used (this should be visible in the spontaneous emission properties). However, no chiral emission is observed at low excitation density. The observed chirality at high fluences could arise from propagation effects (several um-tick samples) present above the threshold but absent for spontaneous emission.
8. The proposed theoretical approach is elegant (nice work!) and qualitatively reproduces the experimental results.

However, the model fails to account for certain specific effects that may be present in the experiments, such as propagation and stimulated emission, and does not provide an independent verification of the underlying photophysics.
For these reasons, I cannot recommend the manuscript for publication.

Referee #2

(Remarks to the Author)
In this manuscript, Wei et al. report circularly polarized superfluorescence from L2MAn−1PbnI3n+1 (n = 3). Circular polarization is induced by chirality in the structure due to organic spacers.
Polarization-resolved measurements clearly show that the SF emission is circularly polarized based on the chirality of the spacer material. The theory supports the experimental results. The paper also indicates that an external magnetic field can tune the effect. The observation of chiral superfluorescence is a significant result as it connects two emerging fields in condensed matter: chirality-induced electronic and optical properties and solid-state superfluorescence. Thus, these findings are significant.

However, the experimental analysis related to SF characterization lacks essential details. Some crucial discussions regarding interpreting the experimental data are presented too briefly. There also appear to be some inconsistencies between different experiments. These details and inconsistencies need to be addressed before publication in any journal.

1- The data in Figure 2a clearly show that the PL signal exhibits pronounced ringing. This is one of the data that is consistent with earlier SF reports in similar systems. However in their intensity analysis, they correlated the peak intensity of the SF transients with the excitation fluence. I am not sure if the exciton density is linearly correlated with the excitation fluence for this extensive range of excitation fluences. At low fluences, the population density can increase linearly; as the density increases, it will saturate.
2- They probed population kinetics using transient reflection spectroscopy around 630 nm. But SF is taking place at 705 nm. They do not have much reflection signal at the relevant wavelengths. However dynamics related SF should be measured at that wavelength.
3- Are the data in Figures 2c and 2b correlated? There are six delay values in Figure 2c and 7 PL traces in 2b. It looks like the lowest fluence is 45 micro J in Figure 2c. The delay observed at this point is 10s of ps. However, the transient reflection experiments show 25micro J data and provide a delay of around 5 ps. According to PL experiments, at 25 micro J, excitation is below the threshold.
4- In discussing the time-resolved reflection data (154 microJ excitation), they refer to the very sharp signal around zero delay as the SF burst. However, that does look like a feature in all experiments starting from 25microJ. While its intensity depends on the excitation power, it has the same narrow transient behavior. From the 2D images, it looks like the cross-correlation signal of the white light continuum with the probe pulse. These types of signals are also known as coherent artifacts in ultrafast optics. If one carefully analyzes the 2D (time/spectra) images, the blue part of that sharp feature comes at earlier delays than the red part. One can use it to correct the chirp in their experiments. That is because, due to normal dispersion, the blue part of the white light arrives in the sample later than the red. Hence, when the probe is scanned from negative delay (longer path) to positive delay (shorter path), the cross-correlation of the white light with the probe reveals the chirp. In short, this feature is not an SF burst.

5- In lines 130-135, they first describe transient reflection experiments and introduce the sharp feature as the SF burst is. But then they start talking about ringing dynamics in 300ps. They probably refer to the PL data at this point. This transition is not clear. But assuming that they refer to the PL data. This discussion suggests the two time-resolved experiments are correlated. If so the bursts in the two different experiments should have a correlated time duration. Yet the burst in the transient reflection is more than an order of magnitude shorter compared to the burst in PL. I would expect the population dynamics to show steady, slow-decaying dynamics and very sharp decay. And the decay time of the population to correlate with the burst time observed in the time-resolved PL experiments.

6- At the end of the same paragraph, they mention dynamic redshift in the spectra, which has no connection to the rest of the discussion.

Overall, the SF analysis section is very brief. They need to provide a more detailed analysis and discussion of these results and clarify these ambiguities.

Referee #3

(Remarks to the Author)
This manuscript repots circularly polarized superfluorescence from chiral perovskite quantum well superlattices. The finding is the first of its kind and could generate a lot of excitement in the field. It could therefore be suitable for a publication at Nature. Authors, however, need to clarify the following points.
1. Authors need to explain why it is necessary to grow “crisscross network” of quantum well superlattices. Ref. 33 report circularly polarized PL from microplates of a similar chiral perovskite material. Would that be possible to grow simple microplates.
2. Superlattices of PEA and RMBA in Supplementary Fig. 2 show a significantly different density and morphology. A similar zoom in figure should be shown for SMBA. The structure and size of the superlattices are at the same length scale as the

wavelength of the light. This leads me to wonder whether the photonic effects have any influence on the observed behavior. A circular dichroism spectrum on the sample would be helpful in understanding this.
3. Ref. 33 reported observation of circularly polarized PL at low temperature. Do the samples of this manuscript show circularly polarized PL at low temperatures?
4. Authors show that DOP only increase above the SF threshold. But below the threshold PL band is broad and weak. Could this make the DOP measurement difficult.
5. If the CP is observed only in SF stage, author need to explain why.

Version 1:

Reviewer comments:

Referee #1

(Remarks to the Author)
The authors have substantially revised the manuscript and provided extensive new experiments, along with the development of a more comprehensive theoretical framework that addresses many of the concerns raised in the previous referee report. Of particular importance are the experiments yielding very long coherence time (up to 70 ps) at room temperature. If true, this result is truly remarkable for solid-state excitons and, to the best of my knowledge, may represent the first report of such long coherence for bare excitons at room temperature.
Given the novelty and significance of this finding, a more in-depth analysis is required. In particular, a solid theoretical explanation is needed to clarify why the edge states exhibit such exceptional coherence properties. The authors attribute this behaviour to carrier localization; however, localization typically enhances exciton–phonon coupling by relaxing momentum conservation. Moreover, edge states are expected to be more localized at the surface, which should further strengthen interactions with the organic spacer ligands, thus enhancing disorder-induced dephasing and emission broadening. This raises a fundamental question: why is the coherence time so long in this case? Why is the exciton–phonon coupling so different from that of bulk excitons in 2D layers? A detailed understanding of these aspects is crucial.
Moreover, edge states have been shown to strongly depend on environmental factors, with moisture significantly affecting their PL QY. This makes the reported long exciton coherence somewhat counterintuitive and unexpected. How robust is the PL from these edge states? How was the sample treated? These details are crucial for further experimental validation and reproducibility.
In addition, further simulations are required to conclusively support the interpretation of the directional emission measurements. The peculiar crisscross network structure could hinder long-range propagation. In such a scenario, amplified spontaneous emission (ASE can occurs on the hundreds um-scale length) may be more efficiently scattered out of plane, potentially explaining the enhanced emission observed at normal incidence.
Without addressing these important insights, the manuscript does not yet reach the quality level required for publication in Nature.

Referee #2

(Remarks to the Author)
I appreciate the authors' meticulous and comprehensive response to my previous report. In the revised manuscript and supplementary materials, they have significantly strengthened the analysis and addressed the key concerns I raised regarding the identification and interpretation of superfluorescence.

The revision now presents multiple, consistent lines of evidence supporting the attribution of the observed emission to superfluorescence rather than to coherent artifacts, amplified spontaneous emission, or photonic effects. These include fluence-dependent scaling and delay-time behavior aligned with SF theory, clear experimental differentiation from ASE through directionality measurements and controls, time-resolved signatures of collective emission, and independent coherence measurements confirming that the relevant timescales for SF are met.

The authors have also clarified the role of sample geometry and ruled out trivial photonic or waveguiding origins of the observed polarization and directionality. While some aspects of the chiral, forward-directed emission are discussed at a qualitative level, they are clearly framed as interpretations and do not affect the central identification of superfluorescence.

Overall, the manuscript is now internally consistent, well supported experimentally, and significantly improved. I believe the authors have adequately addressed my earlier concerns and support publication in its present form.

Referee #3

(Remarks to the Author)
Authors have performed multiple new experiments as well as theoretical modeling to address the comments of all the reviewer. These new experiments and theory work address all the key concerns of the reviewer and substantially improved the paper. Now I believe that the paper provide not only a strong evidence of chiral SF but also a plausible theoretical explanation. This work represents a significant advance of the field worthy of publication in nature. I can now recommend the paper as it is.

Version 2:

Reviewer comments:

Referee #1

(Remarks to the Author)
The authors have clarified their views on long exciton coherence times. Without further technical comments, I would support publishing this version.

Referee #2

(Remarks to the Author)
The authors have revised the manuscript and have addressed the concerns raised by Referee #1. The coherence properties, including the dephasing time, are sufficient to support the observation of superfluorescence, as the condition that the coherence time exceeds the buildup time is satisfied. The addition of four-wave mixing measurements provides a direct and independent confirmation, further strengthening the experimental basis. Together with the threshold behavior, superlinear scaling, delay dynamics, and directionality, the identification of superfluorescence is well supported.

The main advance of the work, namely, the demonstration of chiral superfluorescence and its control via material handedness and magnetic field, is clearly established. The emergence of circular polarization, its sign reversal, and its tunability are unambiguous and represent a significant result, supported by a consistent theoretical description.

The question regarding the origin of the long coherence time pertains to a deeper microscopic understanding rather than the validity of the central claim. The authors provide a physically reasonable explanation grounded in prior literature, which is sufficient for the scope of the present work. In my view, the points raised by Referee #1 have been adequately addressed.

Regarding statistical analysis, the fitting procedures and reported uncertainties are appropriate for the type of measurements presented. Error bars are clearly defined where relevant, and no additional statistical treatment is required.

Overall response to all reviewers

We sincerely thank the reviewers for their feedback, which has been invaluable in strengthening our manuscript. We have thoroughly addressed all comments by incorporating substantial new experimental data, detailed theoretical analyses, and expanded discussions that significantly enhance the study's rigor and clarity.

The major improvements in our revised manuscript and supplementary information include:

1) **Robust demonstration of SF from edge states:** Comparative studies with exfoliated microplates (PL, PL mapping, TRPL), combined with optical simulations and DFT calculations, establish that the SF emission originates from edge states within our superlattices (Extended Data Fig. 5, Supplementary Note 4, Supplementary Figs. 3–6).
2) **Direct evidence of long coherence time:** New transient four-wave mixing (FWM) spectroscopy measurements reveal a long coherence dephasing time (T_2) for edge states at room temperature, providing the fundamental basis for observing room-temperature SF (Supplementary Note 8, Supplementary Fig. 17).
3) **Confirmation of coherent coupling:** Additional TRPL studies demonstrate clear signatures of coherent dipole interactions (Supplementary Note 5, Supplementary Figs. 7-10).
4) ***N*² scaling of collective emission:** Additional data points in power-dependent TRPL spectra with improved fits confirm the characteristic quadratic dependence of SF intensity on excitation density ($I_{SF} \propto N^2$), a fundamental signature of SF (Fig. 2, Extended Data Figs. 6-7).
5) **Artifact-free population dynamics:** New pump-probe measurements in transmission geometry, conducted on transferred superlattices, provide clean transient absorption data that clearly show the characteristic SF population dynamics and delay time ($\tau_D \propto \ln N/(N+1)$), matching theoretical predictions and perovskite SF literature (Fig. 2, Supplementary Fig. 15).
6) **Unambiguous distinction from ASE:** New directional emission studies confirm that the observed SF emission is distinct from ASE, as the signal is strongest in the forward direction rather than along the longer optical path. The temporal dynamics (Burnham-Chiao oscillations, delayed emission) further confirm SF behavior (Supplementary Note 6, Supplementary Figs. 12-13).
7) **Mechanism of circular polarization amplification:** New quantitative quantum optics calculations reveal that DCP amplification in the SF regime arises from collective chiral dipoles, which agree well with experimental results (main text section “Circularly polarized chiral superfluorescence”, Methods, Fig. 3d).

These revisions include new data in Figs. 2, 3, Extended Data Figs. 4-5, and Supplementary Notes 4-8, with Supplementary Figs. 3-10, 12, 15-17, 19-22, 24. Below we provide detailed point-by-point responses to each of the reviewers’ comments.

Reviewer texts is in red.
Major revisions in Main Text and SI are marked in blue.
References cited in the responses are listed at the end of each reply.

Response to the reviewers' comments

Referee #1 (Remarks to the Author):

The authors report the realization of strong left- and right-handed circularly polarized superfluorescence (CP-SF) from large-area (>100 μm × 100 μm) vertically aligned chiral perovskite quantum well superlattices at room temperature. Supported by theoretical quantum optics calculations, they claim that chirality-induced photon transport facilitates the transition to CP-SF from initially incoherent, unpolarized spontaneous emission, resulting in an amplified degree of circular polarization of up to approximately 14%. The application of a low external magnetic field can further modulate the degree of CP-SF.

Achieving high-temperature cooperative emission is an extremely attractive goal, with the potential to unlock quantum photonic applications by reducing system complexity and eliminating the need for cryogenic apparatus. In addition, the claimed observation of circularly polarized SF could stimulate further research in the fields of quantum optics and spintronics. However, the main claim, the observation of SF at room temperature, presents a fundamental challenge: how to retain the coherence properties of excitons in solid-state compounds can be retained and protected against decoherence processes at elevated temperatures. Despite its promise, the current manuscript fails to provide robust evidence to substantiate the very main claim.

Response:

We thank the reviewer for pointing out the significance of our work and for highlighting the fundamental challenge of achieving room-temperature superfluorescence (RT-SF). Through an extensive set of additional experiments (including sample transfer for pump-probe measurements, synthesis of control microplates, coherent coupling studies, and direct dephasing-time measurements) we now have robust evidence to substantiate our claim through multiple, consistent lines of investigation.

Our observation of RT-SF is supported by key signatures that align with established SF criteria:

- **Quadratic Scaling of Peak Intensity:** With the addition of more data points in the revised manuscript, we now clearly demonstrate the quadratic scaling of SF peak intensity with excited dipole density ($I_{SF} \propto N^2$). This was determined by first establishing the scaling between excited dipole density and pump fluence in the spontaneous emission regime ($N \propto P^{\alpha}$). The subsequent quadratic scaling of the SF intensity ($I_{SF} \propto P^{2\alpha}$) directly yields $I_{SF} \propto N^2$ (Fig. 2b, Extended Data Figs. 6–7).
- **Delay Time and Ringing Dynamics:** Our determination of $N \propto P^{\alpha}$ also allows us to verify the theoretically predicted scaling of the SF delay time ($\tau_D \propto \ln N/(N+1)$). This scaling is now confirmed through two independent lines of evidence: TRPL measurements and new transient absorption (TA) spectroscopy measurements (Fig. 2d). Moreover, our TRPL spectra show clear evidence of Burnham-Chiao ringing (Figs. 2a and 2b), another hallmark property of SF.
- **Exclusion of ASE:** New directional emission measurements definitively rule out amplified spontaneous emission, as the signal is strongest in the forward direction, contrary to the path-length-dependent gain expected for ASE (Supplementary Note 6, Supplementary Figs. 12, 13).

- **Identification of SF from Edge States** (**Supplementary Note 4**)**:** Crucially, we have also identified the mechanism that enables coherence preservation at room temperature: The SF at ~690-710 nm originates from **localized edge states** within our superlattices, as confirmed by comparative studies on single-crystal microplates. We have made the following key new measurements:
    - **Direct Measurement of Long Coherence (Supplementary Note 8):** New transient four-wave mixing (FWM) spectroscopy directly reveals a long coherence dephasing time (T_2) of up to ~70 ps at room temperature. Even at high pump fluences, T_2 remains longer than the SF delay and pulse duration, satisfying the fundamental condition for SF.
    - **Suppressed Decoherence (Supplementary Note 4):** Temperature-dependent measurements show minimal emission linewidth broadening (~10 meV from 77 K to 300 K, Fig. 7), starkly contrasting the significant broadening seen in bulk excitons. This indicates dramatically reduced electron-phonon coupling, allowing for coherence to persist.

In summary, the combined verification of theoretically predicted SF scaling laws, the exclusion of ASE, and the identification of long-lived, localized edge states as the emitting species provide robust evidence for RT-SF. Following the reviewer's suggestions, we have incorporated these new data and discussions throughout the revised manuscript to firmly address this central claim.

Here are more detailed comments:

1. There is no direct experimental proof of exciton coherence, which is crucial when invoking coherent coupling.

Response:

We thank the reviewer for raising this crucial point. To directly address the need for experimental proof of exciton coherence, we have performed two new sets of experiments.

First, we conducted transient four-wave mixing (FWM) spectroscopy, which directly measures the dephasing time T_2 of the edge-states (as answered above, SF is from edge states). The results confirm a long T_2 at room temperature, establishing the prerequisite phase memory for coherent coupling (Supplementary Note 8, Supplementary Fig. 17).

Second, as an independent line of evidence, we measured the spontaneous decay dynamics under quasi-resonant versus non-resonant excitation at low pump fluence (Supplementary Note 5). The observed enhancement of the radiative decay rate under quasi-resonant conditions at the SF wavelength is a clear signature of coherent coupling, indicating the formation of phase-aligned dipoles that can interact with a common radiative mode (see also Ref. 18 of the SI).

Together, the direct measurement of a long T_2 via FWM and the observation of excitation-condition-dependent cooperative decay provide robust and complementary evidence for the emitter coherence essential for SF. These new data are presented in Supplementary Note 5, Supplementary Note 8, and Supplementary Figs. 7–10 and 17.

2. Oscillations in the time domain are not exclusively indicative of superfluorescence (SF). They can also arise from peculiar intraband relaxation in stimulated emission experiments, as

discussed in J. Phys. B: At. Mol. Opt. Phys., 39, 215 (2006), and J. Appl. Phys., 91, 6367 (2002). A more detailed analysis is needed to rule out the amplified spontaneous emission (ASE) process.

Response: We agree that temporal oscillations in SF require careful distinction from ASE. Our revised manuscript provides new experimental evidence that decisively rules out ASE. In particular, a hallmark feature of ASE is stronger emission along the longer optical path (Ref. 1 below, see citations at the end of this response). By contrast, our new directional measurements show that the SF from our superlattices is always strongest in the forward direction (normal to the substrate). This is in stark contrast to our control measurements performed on $CsPbBr_3$ nanocrystal thin film, which exhibits prototypical ASE with amplified gain at the ends of a stripe excitation (Supplementary Figs. 12-13). Furthermore, the ASE oscillations cited by the reviewer rely on very slow energy transfer and state repopulation. Our time-resolved PL and pump-probe data show no evidence of such processes, making that ASE mechanism unlikely for our system.

Please also note that the oscillations exhibited by our superlattices follow the established time evolution of SF, where each burst is well-described by a $sech^2$ pulse (Supplementary Note 1). This provides clear evidence of Burnham-Chiao ringing that closely matches previously published results on SF in cryogenic perovskite systems (Refs. 1,2, Fig. R1 below). Additionally, our TRPL and new transient absorption measurements show the correct theoretical scaling for the delay time of these oscillations (Fig. 2d)—a key signature of SF—which is consistent with previous observations of SF in perovskites at 78K (Fig. R2 below, Ref 3). These multi-faceted analyses robustly demonstrate our observations as SF.

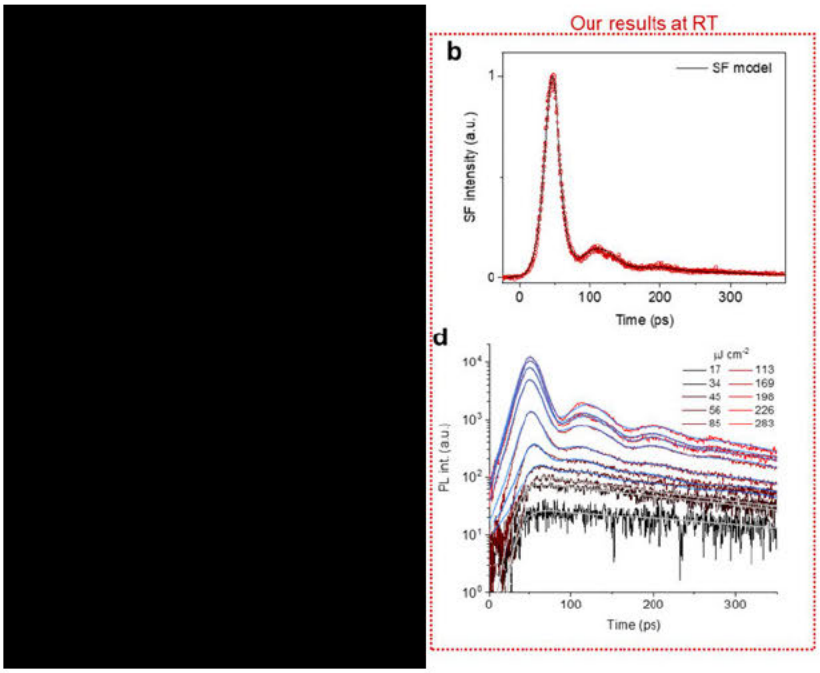

Fig. R1. Comparison with literature for TRPL results. **a**, Characteristics of SF dynamics after pulse excitation (Ref [2]). **b**, Our SF from SMBA perovskite superlattice at room temperature under 198 μJ cm^{-2} pump fluence. **c**, Reported time-resolved PL traces under different pump

fluence of perovskite quantum dots at 6 K (Ref [3]). **d**, Our time-resolved PL traces under different pump fluence of from SMBA perovskite superlattice at room temperature. The light blue lines for SF dynamics are fitting with SF model (Supplementary Note 1). The gray line is the single-exponential decay fitting for spontaneous emission below SF threshold.

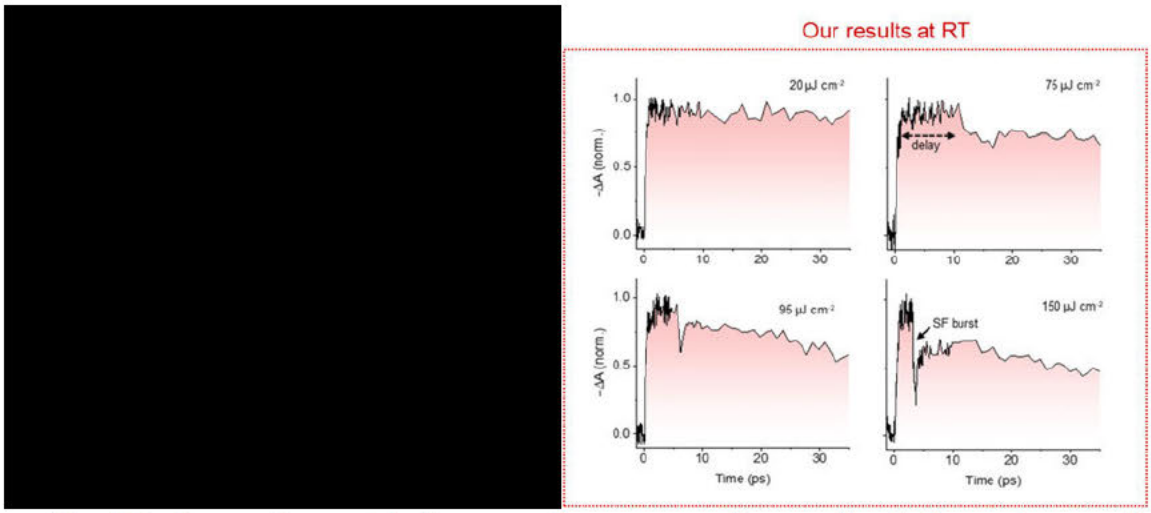

Fig. R2. Comparison with literature for pump-probe results. Left panel: normalized transient absorption dynamics of $MAPbI_3$ film at 78 K (Ref [4]). Right panel: normalized transient absorption dynamics of our SMBA perovskite superlattice at room temperature.

Ref:

1 Saleh, B. E. MC Teich, Fundamentals of photonics. *Ch* 14, 832 (1991).

2 Noe, G. T. *et al.* Giant superfluorescent bursts from a semiconductor magneto-plasma. *Nat Phys* 8, 219-224, doi:10.1038/Nphys2207 (2012).

3 Rainò, G. *et al.* Superfluorescence from lead halide perovskite quantum dot superlattices. *Nature* 563, 671, doi:10.1038/s41586-018-0683-0 (2018).

4 Findik, G. *et al.* High-temperature superfluorescence in methyl ammonium lead iodide. *Nature Photonics* 15, 676-680, doi:10.1038/s41566-021-00830-x (2021).

3. The claimed 14% degree of circular polarization is significantly lower than the values recently reported for chiral ASE and lasing (see for example, https://doi.org/10.1002/adma.202301573, https://pubs.acs.org/doi/full/10.1021/acs.nanolett.4c03838). What could be the benefit of CP-SF?

Response:

We appreciate the reviewer's question. The cited works achieve high polarization ASE/lasing of perovskite nanocrystals using external chiral photonic structures (cholesteric liquid crystals) as reflectors, which selectively reflect right- or left-handed CPL to achieve polarization selectivity. In contrast, our 14% coherent circular polarization arises *intrinsically* from the material's electronic structure via the SF process. This intrinsic chiral SF offers fundamental advantages:

1) **Simplified Architecture:** Our system is a single, solution-processed material. It requires no complex integration of external resonators or photonic structures, significantly simplifying fabrication.

2) **Inherently Chiral Gain Medium:** The chirality is embedded within the gain medium itself, not imposed by an external filter. This eliminates constraints related to spectral alignment with a separate resonator and offers a more direct path to engineering chiral light sources.
3) **Quantum Optical Phenomenon:** The SF process itself provides a unique benefit: the observed DCP is not merely inherited from the spontaneous emission but is *amplified* by the collective quantum dynamics, demonstrating a novel pathway to generating polarized light.
4) **Application Potential:** The intrinsic nature and ultrafast dynamics of chiral SF are highly compatible with future electrically pumped devices and ultrafast coherent chiral opto-spintronic applications, where external liquid crystal layers would be prohibitive.

Therefore, the significance of our work lies in demonstrating a new *mechanism* for generating coherent circularly polarized light—room-temperature chiral SF from an intrinsic chiral gain medium—opening a distinct and promising avenue for quantum optics and chiral photonics.

4. The reported 1 mm-long spatial coherence seems incompatible with a single SF domain. This is more likely to originate from a lasing/photonic mode.

Response:

We thank the reviewer for this important comment. We agree that a 1 mm spatial coherence length would be incompatible with a *single* SF domain (i.e., a single, contiguous, phase-locked giant dipole). We wish to clarify that our observations instead reflect the high spatial coherence of the *emitted light field* from a macroscopic array of coupled SF domains. The ~1 mm spatial coherence was directly measured from the interference fringes in a Michelson interferometer (Supplementary Fig. 14), confirming a high phase correlation across the emission source. This is distinct from the temporal coherence length, which is calculated from the spectral linewidth as $L_C = \lambda^2/\Delta\lambda$ to be ~50 μm. This ~50 μm length is the relevant scale for the SF process itself. Crucially, this coherence length is significantly larger than the spacing between individual emitters in our superlattice, allowing them to interfere and 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. Critically, the absence of sharp cavity modes and the verification of theoretically predicted scaling laws for SF (e.g., quadratic peak intensity, delay time, Burnham-Chiao ringing) definitively rule out lasing or photonic modes.

In summary, the ~50 μm temporal coherence enables the underlying SF process within each collective domain, while the ~1 mm spatial coherence reflects the exceptional macroscopic uniformity of the collective emission from our entire superlattice. We have clarified this distinction in Supplementary Note 7.

5. The transient peak intensity at 705 nm increases superlinearly with pump fluence, with an exponent greater than 2. This behavior is inconsistent with SF buildup and is more likely associated with stimulated emission. Although the authors invoke bimolecular recombination (which is less likely in these compounds due to strong quantum confinement and large binding energies), a detailed analysis to support this claim is missing.

Response:

We thank the reviewer for this comment. The apparent discrepancy arises from the unique photophysics of our emitting species—the edge states—which exhibit a superlinear dependence of the spontaneous emission (SE) intensity on pump fluence even below the SF threshold.

Our revised results (now with more data points and improved fittings) show that the SE intensity at low fluence scales as $I_{SE} \propto P^{1.5\text{-}1.6}$ (for SMBA in Fig. 2b, for RMBA/PEA in Extended Data Figs. 6 & 7). This signifies that the excited dipole density N scales superlinearly with pump power ($I_{SE} \propto N \propto P^{1.5\text{-}1.6}$), a behavior characteristic of these specific edge states in quasi-2D perovskites (consistent with Ref. 1, see Fig. R3a below). This intermediate scaling is understandable for the n=3 structure, which lies between the purely excitonic ($I_{SE} \propto P$) and bimolecular ($I_{SE} \propto P^2$) regimes. We note that the weak bulk exciton emission at ~600 nm only appears under very high fluence above the SF threshold (Fig. R3b below, Supplementary Fig. 3 in the SI), and its intensity scales linearly with pump power—in clear contrast to the superlinear dependence observed for the edge states.

Critically, the superfluorescence intensity is expected to scale quadratically with the number of coupled dipoles ($I_{SF} \propto N^2$). Therefore, the observed pump fluence dependence is a compound effect: $I_{SF} \propto N^2 \propto (P^{1.55})^2 \approx P^{3.16}$ (for SMBA in Fig. 2b); $I_{SF} \propto N^2 \propto (P^{1.51})^2 \approx P^{2.96}$(for RMBA in Extended Data Fig. 6); $I_{SF} \propto N^2 \propto (P^{1.49})^2 \approx P^{2.88}$(for PEA in Extended Data Figs. 7).

This explains why the pump dependence has an exponent greater than 2. The relationship $I_{SF} \propto N^2$ holds, consistent with SF, while the higher-order scaling of N with P is an intrinsic property of the edge-state population mechanism. We note that the smaller exponents originally reported ($I_{SF} \propto P^{2.5\text{-}2.6}$) in our previous submission resulted from inadvertently including data in the high-fluence saturation regime, which has been corrected in the revised fitting.

We have clarified this analysis in the revised main text section "Superfluorescent characteristics and dynamics", the Methods, as well as in Supplementary Note 4.

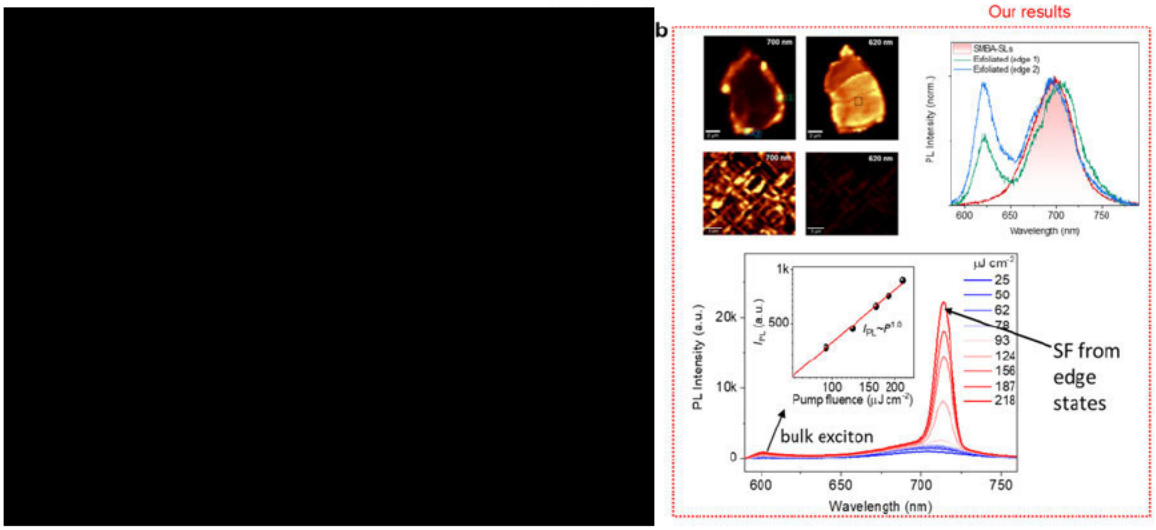

Fig. R3. a, Top left: PL mapping of a $BA_2MA_{n-1}Pb_nI_{3n+1}$ (n = 3) single exfoliated crystal. Top right: Comparison of the PL spectra of the exfoliated crystal, at the exfoliated-crystal edges, and in the corresponding thin film. Bottom left: Schematics of the edge states in

$BA_2MA_{n-1}Pb_nI_{3n+1}$ (n = 3) thin-film perovskite. Bottom right: pump power dependent PL intensity showing superlinear pump dependence of edge states, compared to linear scaling of the bulk. LES: layer edge states (Ref. 1). **b**, Top: PL mapping and PL spectra of exfoliated single crystal flake vs SMBA-SL at low pump fluence. Bottom: pump fluence dependent PL spectra of SMBA-SL, the weak bulk exciton emission bands at ~ 600 nm from quasi-2D perovskite is only observed under extremely high pump fluence.

Ref:

1. Blancon, J.-C. *et al.* Extremely efficient internal exciton dissociation through edge states in layered 2D perovskites. *Science* 355, 1288-1292 (2017). https://doi.org/doi:10.1126/science.aal4211

6. The change in decay time as a function of excitation density is rather abrupt, as expected for the transition from spontaneous to stimulated emission. Please note that the 120 ps decay time at room temperature is likely largely dominated by non-radiative processes.

Response:

We thank the reviewer for this comment. As correctly noted by Reviewer 2, the sharp feature in our original transient reflection data was a coherent artifact. To resolve this, we developed a new sample transfer process (Supplementary Fig. 15) and reconfigured our setup for artifact-free transient absorption (TA) in transmission with cross-polarized pump and probe beams.

The revised TA data, probed directly at the SF wavelength, now clearly show the characteristic signatures of SF:

- Below the SF threshold, we observe a rapid population buildup followed by a stable plateau.
- Above threshold, a distinct delay period (τ_D) emerges before a rapid, collective decay of the population.
- This delay time systematically shortens with increasing excitation density, following the predicted SF relationship $\tau_D \propto \ln N/(N+1)$, consistent with our TRPL results (Fig. 2c). The observed dynamics bear striking resemblance to previous reports of SF in $MAPbI_3$ perovskite thin films at 78 K (Ref. 3), despite the significant difference in measurement temperature (Fig. R2, above).

These dynamics—specifically the presence of a density-dependent delay preceding the abrupt decay—are hallmark signatures of SF, arising from the mutual synchronization of dipoles. They are strongly consistent with measurements of SF reported in other perovskite systems at cryogenic temperatures (Ref 2, 3). The consistency between our TA and TRPL data provides additional independent lines of support for this interpretation over stimulated emission.

Ref:

2 Noe, G. T. *et al.* Giant superfluorescent bursts from a semiconductor magneto-plasma. *Nat Phys* 8, 219-224, doi:10.1038/Nphys2207 (2012).

3 Rainò, G. *et al.* Superfluorescence from lead halide perovskite quantum dot superlattices. *Nature* 563, 671, doi:10.1038/s41586-018-0683-0 (2018).

7. It is unclear why spontaneous emission is not circularly polarized, as reported for similar

compounds (https://www.nature.com/articles/s41467-020-18485-7, https://pubs.acs.org/doi/10.1021/acsnano.9b00302). As claimed by the authors, there should be net crystallographic helicity due to the chiral ligand used (this should be visible in the spontaneous emission properties). However, no chiral emission is observed at low excitation density. The observed chirality at high fluences could arise from propagation effects (several um-tick samples) present above the threshold but absent for spontaneous emission.

Response:

We thank the reviewer for raising this important point regarding circular polarization. We have conducted new, more sensitive measurements that clarify the polarization behavior, and we have developed a new theoretical formalism that explains the transition from unpolarized spontaneous emission to circularly polarized chiral SF.

Experimentally, improved measurements with a higher signal-to-noise ratio show a small but finite DCP in the spontaneous emission at room temperature, measuring ~0.2% (Supplementary Fig. 22). This magnitude is consistent with reports for similar chiral perovskites using the same chiral ligands, where the DCP is typically very small (~10^{-5}-10^{-3}) at room temperature (Refs. 1-3 below). For instance, one of the cited references by the reviewer (https://pubs.acs.org/doi/10.1021/acsnano.9b00302) explicitly states that the DCP "*almost vanishes at room temperature*" in their microplates. Regarding the references cited by the reviewer, we also note that Ref. cited (https://www.nature.com/articles/s41467-020-18485-7) did not actually investigate circularly polarized emission.

Moreover, we have now developed a comprehensive theory that explains why the spontaneous emission at low pump fluence is unpolarized, whereas SF at high pump fluence is circularly polarized. Details are provided in the main text section “Circularly polarized chiral superfluorescence” and in the Methods section “Theoretical modeling of chiral superfluorescence”, which we briefly summarize here. Our chiral superlattices crystalize into a 2_1 screw axis parallel to the direction of SF emission (Supplementary Fig. 18). The dipole orientations consistent with this screw axis are described by Eq. (1) in the main text and shown schematically in Fig. 3a. They are linearly polarized in the plane perpendicular to SF emission and rotate along the screw axis. When excited independently, these dipoles produce an unpolarized radiation pattern. By contrast, the spontaneous generation of circular polarization results from the *collective* excitation of an ensemble of these helically aligned dipoles. As described in Eq. (3), the presence of collective excitations generates a **chiral photonic spin-orbit coupling**, leading to circularly polarized SF.

When the influence of room-temperature dephasing is accounted for, the results of this theoretical model show excellent agreement with our experimental measurements (Fig. 3d). At low pump fluence, the dephasing rate γ_ϕ is much larger than the initial emission rate $I(0) = N\Gamma$ and no coherence is formed to produce SF. In this regime, the dipoles radiate independent, unpolarized spontaneous emission. At high pump fluence, the dephasing rate is negligible and the collective, circularly polarized dynamics dominate.

Furthermore, we have ruled out the possibility that this amplification stems from propagation

effects in our thick samples. Our photonic simulations (Extended Data Fig. 4j) confirm that the isotropic "crisscross" structure of our superlattices shows no inherent preference for amplifying one circular polarization over the other through propagation. This is corroborated by the complete absence of both CD signal and chiral SF in our achiral PEA superlattices, which possess a similar morphology (Figs. S19, S22).

In summary, the circular polarization is very weakly present in spontaneous emission at room temperature, but its significant amplification under SF conditions is an intrinsic quantum optical effect of the collective state, not a structural propagation artifact.

Ref:

1. Ma, J. Q. *et al.* Chiral 2D Perovskites with a High Degree of Circularly Polarized Photoluminescence. *ACS Nano* **13**, 3659-3665 (2019).
2. Li, Hao, et al. "Enhancing Chiroptoelectronic Activity in Chiral 2D Perovskites via Chiral–Achiral Cation Mixing." *Adv Opt Mater* 12.35 (2024): 2401782. (Page S20 of SI)
3. Cao, Q. X. *et al.* Chiral Perovskite Nanoplatelets with Tunable Circularly Polarized Luminescence in the Strong Confinement Regime. *Adv Opt Mater* **11** 2203125 (2023). (Fig.5).

8. The proposed theoretical approach is elegant (nice work!) and qualitatively reproduces the experimental results. However, the model fails to account for certain specific effects that may be present in the experiments, such as propagation and stimulated emission, and does not provide an independent verification of the underlying photophysics.

Response:

We thank the reviewer for their positive assessment of our theoretical approach. In response, we have significantly extended our model into a comprehensive theoretical framework. This framework now quantitatively accounts for the central observation: the transition from nearly unpolarized spontaneous emission at low fluence to circularly polarized SF at high fluence. Our revised theory demonstrates excellent quantitative agreement with the experimentally measured peak degree of circular polarization (DCP_0) across a wide range of pump fluences (Fig. 3d). This strong correspondence provides an independent theoretical verification of the chiral SF mechanism and directly addresses the photophysics underlying the polarization amplification observed in our experiments.

The reviewer is correct that our current model does not include propagation effects, such as photon re-absorption and re-emission responsible for Burnham–Chiao ringing. These non-Markovian effects, which cause deviations from the idealized single-burst SF model, have been extensively studied in seminal works on SF (e.g., Refs. 1,2 below) and do not represent new physics. Because such effects are cumbersome to model analytically, a common approach in the SF literature (including Ref. 3) is to approximate the dynamics as a sum of independent $sech^2$ pulses (Supplementary Note 1), a method we adopt for fitting the SF dynamics in Fig. 2b.

Please also note that the ringing behavior present in Fig. 2b is shown on a logarithmic scale. The same data is shown on a linear scale in Supplementary Fig. 16a, as well as in Extended

Data Fig. 6a for the PEA superlattices and Extended Data Fig. 7a for the RMBA superlattices. The initial SF burst is much larger than the subsequent bursts, indicating that the influence of propagation, absorption, and re-emission is small.

In summary, our theory focuses on the spontaneous emergence of circular polarization—a novel aspect of chiral SF—within a simplified single-burst framework. This choice allows for analytical insight and direct comparison with experiment, while the extension to include propagation follows established formalisms (Refs. 1,2) and would not alter the central conclusions regarding polarization generation.

Refs.

1. Bonifacio, R. et al. Cooperative radiation processes in two-level systems: Superfluorescence. *Phys Rev. A* **11** 1507 (1975) https://link.aps.org/doi/10.1103/PhysRevA.11.1507
2. Bonifacio, R. et al. Cooperative radiation processes in two-level systems: Superfluorescence. II *Phys Rev. A* **12** 587 (1975) https://link.aps.org/doi/10.1103/PhysRevA.12.587
3. Biliroglu, M. *et al.* Room-temperature superfluorescence in hybrid perovskites and its origins (vol 16, pg 324, 2022). *Nat Photonics* **16**, 396-396 (2022). https://doi.org/10.1038/s41566-022-00997-x

For these reasons, I cannot recommend the manuscript for publication.

Response:

We sincerely thank the reviewer for their thorough and constructive feedback, which has been instrumental in strengthening our work. In response, we have conducted extensive new experiments, developed a comprehensive theory, and provided substantial additional evidence to address every concern raised. The consistency of our results across multiple techniques, their alignment with established SF theory, and the systematic exclusion of alternative mechanisms collectively provide a robust and compelling case for our conclusions. We are hopeful that the reviewer will find these revisions persuasive and reconsider our manuscript's suitability for publication.

Referee #2 (Remarks to the Author):

In this manuscript, Wei et al. report circularly polarized superfluorescence from L2MAn−1PbnI3n+1 (n = 3). Circular polarization is induced by chirality in the structure due to organic spacers.

Polarization-resolved measurements clearly show that the SF emission is circularly polarized based on the chirality of the spacer material. The theory supports the experimental results. The paper also indicates that an external magnetic field can tune the effect. The observation of chiral superfluorescence is a significant result as it connects two emerging fields in condensed matter: chirality-induced electronic and optical properties and solid-state superfluorescence. Thus, these findings are significant.

However, the experimental analysis related to SF characterization lacks essential details. Some crucial discussions regarding interpreting the experimental data are presented too briefly. There also appear to be some inconsistencies between different experiments. These details and inconsistencies need to be addressed before publication in any journal.

Response: We appreciate the reviewer's positive assessment of our work's significance and their constructive feedback regarding our experimental and theoretical analyses. In response to the comments, we have substantially strengthened the manuscript by addressing each of the reviewer's concerns, as discussed in detail below.

1- The data in Figure 2a clearly show that the PL signal exhibits pronounced ringing. This is one of the data that is consistent with earlier SF reports in similar systems. However in their intensity analysis, they correlated the peak intensity of the SF transients with the excitation fluence. I am not sure if the exciton density is linearly correlated with the excitation fluence for this extensive range of excitation fluences. At low fluences, the population density can increase linearly; as the density increases, it will saturate.

Response:

We thank the reviewer for pointing out this subtlety and we agree with their assessment. As described in our initial response to Reviewer #1 above, and to Reviewer #1's comment 5, we have performed new time-resolved PL measurements to characterize the scaling of the excitation density on pump fluence in both the low fluence (spontaneous emission; SE) and high fluence (SF) regimes.

In the original Fig. 2, only data for the SF regime were shown. Our revised results (now with more data points and improved fittings) show that the SE intensity at low fluence scales as $I_{SE} \propto P^{1.5\text{-}1.6}$ (for SMBA in Fig. 3b, for RMBA/PEA in Extended Data Figs. 6 & 7). This signifies that the excited dipole density N scales superlinearly with pump power ($I_{SE} \propto N \propto P^{1.5\text{-}1.6}$). We also observe that the SF intensity scales as $I_{SF} \propto N^2 \propto (P^{1.55})^2 \approx P^{3.16}$ (for SMBA in Fig. 2b); $I_{SF} \propto N^2 \propto (P^{1.51})^2 \approx P^{2.96}$(for RMBA in Extended Data Fig. 6); $I_{SF} \propto N^2 \propto (P^{1.49})^2 \approx P^{2.88}$(for PEA in Extended Data Figs. 7), thereby reproducing the expected quadratic intensity scaling on excitation density for SF. Complete TRPL traces from low to high pump fluence are now also provided in both linear and logarithmic scales for the SMBA, RMBA, and PEA superlattice samples (Fig. 2b, Supplementary Fig. 16, Extended Data Figs. 6 & 7).

As described in our response to **Reviewer #1's comment 5** above, this superlinear scaling of excitation density with pump fluence is consistent with our identification of edge states as the

emissive species (see also Ref. 1 in response to Reviewer #1's comment 5 above). We note that the smaller exponents originally reported ($I_{SF} \propto P^{2.5\text{-}2.6}$) in our previous submission resulted from inadvertently including data in the high-fluence saturation regime, which has been corrected in the revised fitting.
We thank the reviewer for suggesting a more careful analysis of the pump fluence dependence which has greatly improved the manuscript.

2- They probed population kinetics using transient reflection spectroscopy around 630 nm. But SF is taking place at 705 nm. They do not have much reflection signal at the relevant wavelengths. However dynamics related SF should be measured at that wavelength.

Response:

We thank the reviewer for this feedback, and we agree that probing the population dynamics directly at the SF emission wavelength is the correct approach. In the revised manuscript, we have completely replaced all previous transient reflection data with new transient absorption measurements conducted at the SF wavelength (705 nm for the SMBA superlattices). These measurements were performed in transmission mode on samples transferred to transparent substrates with cross-polarized pump and probe beams, which directly captures the relevant population kinetics and eliminates potential artifacts associated with reflection-based probing and the previous $MAPbBr_3$ single crystal substrate. The new data (now in Fig. 2) unambiguously show the characteristic SF dynamics, including the expected delay time and rapid population decay in the SF regime (see also Fig. R2 in our response to Reviewer #1 above).

3- Are the data in Figures 2c and 2b correlated? There are six delay values in Figure 2c and 7 PL traces in 2b. It looks like the lowest fluence is 45 micro J in Figure 2c. The delay observed at this point is 10s of ps. However, the transient reflection experiments show 25micro J data and provide a delay of around 5 ps. According to PL experiments, at 25 micro J, excitation is below the threshold.

Response:

We thank the reviewer for highlighting this inconsistency. The reviewer is correct that the pump fluences used for the time-resolved PL (TRPL) traces in the original Fig. 2b did not match those plotted for the delay time in the original Fig. 2c. Specifically, the TRPL data at 34 μJ cm^{-2} (corresponding to spontaneous emission) were not included in Fig. 2c, which was initially focused on presenting SF-regime results.
We have revised the figures to ensure full correspondence and clarity:

1. We have provided complete power-dependent TRPL datasets in both linear and logarithmic scales (Fig. 2b, Supplementary Fig. 16, Extended Data Figs. 6–7) and added model fits to all traces.
2. The new Fig. 2c now plots the delay times for the same pump fluences as shown in the TRPL traces in Fig. 2b, with additional delay-time points derived from the new transient absorption (TA) measurements (Fig. 2d).

Regarding the delay-time discrepancy at 25 μJ cm^{-2} in the original dataset, we identified a measurement artifact in the earlier transient reflection data (addressed in the following

comment). We have since replaced all transient reflection data with **artifact-free transient absorption measurements** on newly transferred samples on transparent substrate, confirming that 25 μJ cm^{-2} is indeed below the SF threshold. This result is now fully consistent with the revised TRPL data in Fig. 2c.
We thank the reviewer for these observations, which have significantly improved the rigor and clarity of our data presentation.

4- In discussing the time-resolved reflection data (154 microJ excitation), they refer to the very sharp signal around zero delay as the SF burst. However, that does look like a feature in all experiments starting from 25microJ. While its intensity depends on the excitation power, it has the same narrow transient behavior. From the 2D images, it looks like the cross-correlation signal of the white light continuum with the probe pulse. These types of signals are also known as coherent artifacts in ultrafast optics. If one carefully analyzes the 2D (time/spectra) images, the blue part of that sharp feature comes at earlier delays than the red part. One can use it to correct the chirp in their experiments. That is because, due to normal dispersion, the blue part of the white light arrives in the sample later than the red. Hence, when the probe is scanned from negative delay (longer path) to positive delay (shorter path), the cross-correlation of the white light with the probe reveals the chirp. In short, this feature is not an SF burst.

Response:

We thank the reviewer for pointing out that the sharp feature in our original transient reflection data was indeed an artifact, To correct this critical issue, we have replaced the original transient reflection data with new transient absorption measurements. To make these measurements, we developed a sample transfer process onto transparent quartz substrates (Supplementary Fig. 15) and reconfigured our setup for artifact-free transient absorption in transmission mode with cross-polarized pump and probe beams. The new data reveal clear signatures of SF:

1) Below threshold: Rapid population buildup followed by slow decay
2) Above threshold: Distinct delay period (τ_D) before rapid, collective decay
3) The delay time follows $\tau_D \propto \ln N/(N+1)$, consistent with SF theory and with our independent TRPL results (Fig. 2c)

5- In lines 130-135, they first describe transient reflection experiments and introduce the sharp feature as the SF burst is. But then they start talking about ringing dynamics in 300ps. They probably refer to the PL data at this point. This transition is not clear. But assuming that they refer to the PL data. This discussion suggests the two time-resolved experiments are correlated. If so the bursts in the two different experiments should have a correlated time duration. Yet the burst in the transient reflection is more than an order of magnitude shorter compared to the burst in PL. I would expect the population dynamics to show steady, slow-decaying dynamics and very sharp decay. And the decay time of the population to correlate with the burst time observed in the time-resolved PL experiments.

Response:

We thank the reviewer for pointing out this concern. As discussed above, we have replaced our original transient reflection data (which contained measurement artifacts) with new data obtained via transient absorption spectroscopy. This new data shows excellent agreement with the TRPL results (Fig. 2c). We have also revised the text to ensure the discussion of these

results is clear.

6- At the end of the same paragraph, they mention dynamic redshift in the spectra, which has no connection to the rest of the discussion.

Response:

We agree that this discussion was not relevant and have removed it.

Overall, the SF analysis section is very brief. They need to provide a more detailed analysis and discussion of these results and clarify these ambiguities.

Response:

We thank the reviewer for this important feedback. In addition to the changes described above, we have also substantially expanded the SF analysis section through the following major revisions (please also see our responses to Reviewer #1's comments 1, 2, 5, and 6):

1. **Identification of edge states as the SF source**. We have provided additional comparative microplate studies, DFT calculations, optical simulations and PL spectra/mapping and lifetime analysis (Extended Data. Fig. 5, Supplementary Note 4, Supplementary Figs. 3-6) demonstrating that SF in our samples originates from the edge states of the superlattices.
2. **Comprehensive analysis of exciton coherence.** Our new four-wave mixing spectroscopy and excitation-wavelength-dependent dynamics measurements (Supplementary Note 5, Supplementary Figs. 8-11) show clear evidence of coherent dipole coupling and long dephasing (T_2) times.
3. **Clearer discussion of SF signatures** including:
    - Improved verification of quadratic SF intensity scaling $I_{SF} \propto N^2$ and delay time scaling $\tau_D \propto \ln N/(N+1)$ through the inclusion of additional data points and new transient absorption measurements
    - New directional emission measurements that rule out amplified stimulated emission (ASE) (Supplementary Note 6, Supplementary Figs. 12-13)
    - Detailed comparisons with established SF literature
4. **Expanded theoretical framework** providing a quantitative explanation of the chiral SF mechanism and amplification of circular polarization.

We believe these revisions, in addition to the corrections and replacement of transient reflection data with new transient absorption data (as described above), have significantly enhanced the manuscript. We thank the reviewer for these suggestions.

Referee #3 (Remarks to the Author):

This manuscript repots circularly polarized superfluorescence from chiral perovskite quantum well superlattices. The finding is the first of its kind and could generate a lot of excitement in the field. It could therefore be suitable for a publication at Nature. Authors, however, need to clarify the following points.

Response:

We sincerely thank the reviewer for their positive assessment of our work's significance and their constructive feedback. We have carefully addressed each of the points raised, as detailed below:

1. Authors need to explain why it is necessary to grow “crisscross network” of quantum well superlattices. Ref. 33 report circularly polarized PL from microplates of a similar chiral perovskite material. Would that be possible to grow simple microplates.

Response:

We thank the reviewer for this important question. The "crisscross network" architecture is indeed essential for achieving SF, as it provides the high density of edge states required for this collective phenomenon. Please see our response to Reviewer #1’s comment 5 above, and the Supplementary Notes 4, 8.2 for details on the identification of edge states as the source of SF. While microplates (Ref. 33 in our original manuscript, https://pubs.acs.org/doi/10.1021/acsnano.9b00302) show chiral photoluminescence from bulk excitons, our new comparative studies of similar microplates reveal that SF originates uniquely from edge states in our perovskite superlattices (Extended Data Fig. 5, Supplementary Notes 3,4, Supplementary Figs. 3-6). The vertically aligned crisscross superlattices provide a dramatically higher density of these edge states compared to microplates or conventional polycrystalline films, as evidenced by SEM/TEM imaging (Fig. 1 and Extended Data Fig. 2). Crucially, we observe SF exclusively in our superlattices, not in exfoliated microplates or Ruddlesden–Popper perovskite polycrystalline thin films (Supplementary Figs. 4, 24). This unique morphology therefore enables the high emitter density, long coherence times (via edge-state localization), and coherent coupling necessary for room-temperature SF, as analyzed in Supplementary Note 4.

2. Superlattices of PEA and RMBA in Supplementary Fig. 2 show a significantly different density and morphology. A similar zoom in figure should be shown for SMBA. The structure and size of the superlattices are at the same length scale as the wavelength of the light. This leads me to wonder whether the photonic effects have any influence on the observed behavior. A circular dichroism spectrum on the sample would be helpful in understanding this.

Response:

We thank the reviewer for raising these important points regarding morphology and potential photonic effects. As suggested, we have added a zoomed-in SEM image of the SMBA superlattice to the revised Supplementary Fig. 2, allowing for a direct comparison with the PEA and RMBA structures. We have also measured the circular dichroism spectrum (Supplementary Fig. 19).

Regarding the potential contribution of photonic effects, we performed comprehensive photonics simulations of our superlattices to explicitly characterize the influence of morphology (Extended Data Fig. 4, Supplementary Note 3). In particular, we included an assessment of whether the edge

luminescence could arise from waveguiding of bulk emission. Our photonic simulations show no near-field intensity distribution at the edge for the 700 nm emission, ruling out a waveguide mechanism (Extended Data Fig. 4i). Furthermore, the significant spectral separation (~80 nm) and distinct lifetimes between edge-state and bulk emissions (Extended Data Fig. 5, Supplementary Figs. 3, 4), along with DFT calculations confirming the existence of narrower-bandgap edge states (Supplementary Fig. 5), collectively support the assignment of the edge luminescence to intrinsic electronic edge states.

Regarding the potential contribution of photonic effects to the observed circular polarization, our simulations (Extended Data Fig. 4, Supplementary Note 3) show identical responses for left- and right-circularly polarized light, definitively ruling out photonic contributions. These results are further supported experimentally by the complete absence of circularly polarized emission in achiral PEA superlattices, which share a similar morphology. Together, these results confirm that the circular polarization in SF emission originates from intrinsic chiral quantum optical interactions within the material, not from structural photonic effects.

Please also note that spontaneous emission from our superlattices is essentially unpolarized (Supplementary Fig. 22a,b with DCP of ~0.2±0.1%) at room temperature. This provides strong evidence for a cooperative quantum optical origin of the circular polarization in the SF regime. Importantly, our new comprehensive theory describes this process in detail and shows excellent agreement with the measured data (Fig. 3d, Methods section “Theoretical modeling of chiral superfluorescence)).

3. Ref. 33 reported observation of circularly polarized PL at low temperature. Do the samples of this manuscript show circularly polarized PL at low temperatures?

Response:

We thank the reviewer for this suggestion. We have performed the requested measurement (Supplementary Fig. 22), which confirms that the samples only exhibit circularly polarized PL at low temperature with DCP up to ~5% at 77K. This is consistent with prior literature (Ref. 1 below), as the DCP in such systems originates from spin-polarized carriers whose coherence is readily quenched by thermal phonon scattering at room temperature. This further highlights the novelty of our room-temperature observation of **chiral superfluorescence** with DCP >13%, which arises from a fundamentally different, many-body coherent mechanism.

Ref:

1. Lu, Haipeng, et al. "Spin-dependent charge transport through 2D chiral hybrid lead-iodide perovskites." *Science advances* 5, no. 12 (2019): eaay0571.

4. Authors show that DOP only increase above the SF threshold. But below the threshold PL band is broad and weak. Could this make the DOP measurement difficult.

Response:

The reviewer is correct that the weak, broad spontaneous emission below threshold makes DCP measurement challenging. With improved measurement sensitivity (increased integration time), we now detect a small but finite DCP of 0.2% in spontaneous emission at room temperature (Supplementary Fig. 22). This magnitude is consistent with other chiral perovskite

systems (Ref. 33 in comment 1 and Refs. 1-3 below).

Ref:

1 Ma, J. Q. *et al.* Chiral 2D Perovskites with a High Degree of Circularly Polarized Photoluminescence. *Acs Nano* **13**, 3659-3665 (2019).

2 Di Nuzzo, D. *et al.* Circularly Polarized Photoluminescence from Chiral Perovskite Thin Films at Room Temperature. *Acs Nano* **14**, 7610-7616 (2020).

3 Cao, Q. X. *et al.* Chiral Perovskite Nanoplatelets with Tunable Circularly Polarized Luminescence in the Strong Confinement Regime. *Adv Opt Mater* **11** 2203125 (2023).

5. If the CP is observed only in SF stage, author need to explain why.

Response:

We thank the reviewer for this comment. We have now developed a comprehensive theory that explains why the spontaneous emission at low pump fluence is unpolarized, whereas SF at high pump fluence is circularly polarized. Details are provided in the main text section "Circularly polarized chiral superfluorescence" and in the Methods section "Theoretical modeling of chiral superfluorescence", which we briefly summarize here. Our chiral superlattices crystalize into a 2_1 screw axis parallel to the direction of SF emission (Supplementary Fig. 18). The dipole orientations consistent with this screw axis are described by Eq. (1) and shown schematically in Fig. 3a. They are linearly polarized in the plane perpendicular to SF emission and rotate along the screw axis. When excited independently, these dipoles produce an unpolarized radiation pattern. By contrast, the spontaneous generation of circular polarization results from the *collective* excitation of an ensemble of these helically aligned dipoles. As described in Eq. (3), the presence of collective excitations generates a **chiral photonic spin-orbit coupling**, leading to circularly polarized SF.

When the influence of room-temperature dephasing is accounted for, the results of this theoretical model show excellent agreement with our experimental measurements (Fig. 3d). At low pump fluence, the dephasing rate γ_ϕ is much larger than the initial emission rate $I(0) = N\Gamma$ and no coherence is formed to produce SF. In this regime, the dipoles radiate independent, unpolarized spontaneous emission. At high pump fluence, the dephasing rate is negligible and the collective, circularly polarized dynamics dominate.

Referee #1 (Remarks to the Author):

The authors have substantially revised the manuscript and provided extensive new experiments, along with the development of a more comprehensive theoretical framework that addresses many of the concerns raised in the previous referee report.

Of particular importance are the experiments yielding very long coherence time (up to 70 ps) at room temperature. If true, this result is truly remarkable for solid-state excitons and, to the best of my knowledge, may represent the first report of such long coherence for bare excitons at room temperature. Given the novelty and significance of this finding, a more in-depth analysis is required. In particular, a solid theoretical explanation is needed to clarify why the edge states exhibit such exceptional coherence properties.

Response:

We thank the referee for acknowledging our extensive revisions and for highlighting the significance of our work. We agree that the long excitonic coherence time at room-temperature is an important feature of our experiments and should be further clarified. First and foremost, we would like to point out that our coherence times are actually in excellent agreement with other reports of 78K and room-temperature SF in a similar quasi-2D PEA:$CsPbBr_3$ hybrid perovskite thin films (Refs. 1-2 below). In those references, a credible theoretical explanation in terms of polaron dressing and the formation of solitons has already been presented. We have substantially revised Supplementary Note 8 to more clearly compare our results with those of Refs. 1 and 2. The key aspects are summarized below.

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. 3). 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), 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:$CsPbBr_3$ hybrid perovskite films exhibiting SF in Refs. 1 and 2. Given the close similarities between our observations and those in the Refs. 1 and 2 (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, may also contribute to the long coherence times in our samples. The formation of solitons, which require the excitons to collectively experience the same lattice environment, is also consistent with our observation of edge states localized within a deep potential well as the SF species.

Please also note that a direct comparison between our results and those of Refs. 1 and 2 requires discerning between the effective dephasing time T_2^* (estimated from the SF delay time) and the homogeneous dephasing time T_2 (measured via four-wave-mixing). Refs. 1 and 2 did not explicitly measure T_2. Nevertheless, T_2 is by definition at least as long as T_2^*, and is often substantially longer in the solid-state because T_2^* includes contributions from inhomogeneous broadening. As such, we can infer that the T_2 times relevant to Refs. 1 and 2 are at least as long as 12.5 ps, and possibly much longer, consistent with our measurements.

We have edited both the main text and **revised Supplementary Note 8** to include this discussion and to more clearly cite the long high-temperature coherence times measured in Refs. 1 and 2. We have also clarified the definitions of T_2 and T_2^*, as well as related measurements pertaining to linewidth broadening, to ensure a clear comparison to values reported in the literature.

Refs:

1) Biliroglu, M. *et al.,* "Room-temperature superfluorescence in hybrid perovskites and its origins". *Nat Photonics* **16**, 396-396 (2022)
2) M. Biliroglu, et al., "Unconventional solitonic high-temperature superluorescence from perovskites", Nature 642, 71-77 (2025).
3) Bonifacio, R. & Lugiato, L. Cooperative radiation processes in two-level systems: Superfluorescence. *Physical Review A* **11**, 1507 (1975).

The authors attribute this behaviour to carrier localization; however, localization typically enhances exciton–phonon coupling by relaxing momentum conservation. Moreover, edge states are expected to be more localized at the surface, which should further strengthen interactions with the organic spacer ligands, thus enhancing disorder-induced dephasing and emission broadening. This raises a fundamental question: why is the coherence time so long in this case? Why is the exciton–phonon coupling so different from that of bulk excitons in 2D layers? A detailed understanding of these aspects is crucial.

Response:

As discussed above, the long coherence times measured in our samples are consistent with those measured in similar quasi-2D hybrid perovskites (Refs. 1-2 above). The dominant mechanism responsible for these long coherences times is likely the polaron dressing described in detail in Refs. 1-2.

Nevertheless, the T_2 times measured in our superlattices are longer than those measured in our own quasi-2D perovskite thin film control sample (Supplementary Fig. 18). Indeed, we attribute this difference to the formation of edge states within the superlattices. We agree with the Reviewer that carrier localization (and the associated broadening in momentum space) can, in principle, enhance elastic phonon scattering (e.g., from defects). However, several complementary measurements, together with prior reports, suggest that edge states in hybrid perovskites are overall less susceptible to decoherence than bulk excitons, likely owing to suppressed inelastic phonon-assisted scattering.

Specifically, our measurements show that the increase in PL linewidth from 77 K to room-temperature is significantly smaller in the superlattices (~10 meV) compared with the bulk exciton broadening (~39 meV) (Supplementary Fig. 6a,b; Supplementary Note 4). This weaker temperature-dependent broadening indicates that the edge states are less sensitive to phonon-induced dephasing; The radiative lifetime of the edge states (1.5 ns in the superlattices) is also substantially longer than the lifetime of bulk excitons (0.32 ns, 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. 4, which showed that edge state localization in similar quasi-2D $(BA)_2(MA)_{n-1}Pb_nI_{3n+1}$ hybrid perovskites with $n \geq 3$ 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 loss channels.

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 time[5-9]. For instance, Ref. 9 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 excitons from the bulk into deep edge states 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 polaron dressing described in Refs. 1 and 2, provide a plausible explanation for the long coherence times overall.

The detailed discussion is provided in **revised Supplementary Note 8.4**. We have also added "The observation of long coherence times exceeding 10 ps is consistent with previous reports of high-temperature SF in similar quasi-2D hybrid perovskites and may arise from suppressed exciton-phonon coupling via polaron dressing[8,24]. In our samples, additional protection against decoherence is likely related to exciton localization in deep edge states[27], which are known to suppresses inelastic relaxation pathways[33] (Supplementary Note 8)." in the revised Main text with relevant Refs.

Refs.

4) Blancon, J.-C. *et al.* Extremely efficient internal exciton dissociation through edge states in layered 2D perovskites. *Science* **355**, 1288-1292, (2017).
5) Poltavtsev, S. V. *et al.* Long coherent dynamics of localized excitons in (In,Ga)N/GaN quantum wells. *Physical Review B* **98**, 195315 (2018).
6) Zhao, H., Moehl, S. & Kalt, H. Coherence Length of Excitons in a Semiconductor Quantum Well. *Physical Review Letters* **89**, 097401 (2002).
7) Hegarty, J. & Sturge, M. D. Studies of exciton localization in quantum-well structures by nonlinear-optical techniques. *J. Opt. Soc. Am. B* **2**, 1143-1154 (1985).
8) Siegner, U. *et al.* Optical dephasing in semiconductor mixed crystals. *Physical Review B* **46**, 4564-4581 (1992).
9) Grisard, S. *et al.* Long-Lived Exciton Coherence in Mixed-Halide Perovskite Crystals. *Nano Lett* (2023).

Moreover, edge states have been shown to strongly depend on environmental factors, with moisture significantly affecting their PL QY. This makes the reported long exciton coherence somewhat counterintuitive and unexpected. How robust is the PL from these edge states? How was the sample treated? These details are crucial for further experimental validation and reproducibility.

Response:

We thank the reviewer for raising this important point regarding the environmental stability of the edge state emission. To address this, we have conducted new measurements to assess the photostability of our superlattice samples under various conditions (see the newly added Supplementary Fig. 12).

Specifically, we measured the emission from edge states below and above the SF threshold in a fresh superlattice sample under two different environments at room temperature: in ambient air (relative humidity ~50%) and in a vacuum of ~10^{-3} Pa. These measurements were performed under

550 nm femtosecond pulsed laser excitation (1 kHz repetition rate, same as in the main text) continuously for over two hours. As shown in the newly added Supplementary Fig. 12, the edge states in our superlattices exhibit high photostability when excited below the SF threshold. At high pump fluence above the SF threshold, the SF emission is extremely robust in vacuum. In ambient air, we observed only a slight reduction in emission intensity after two hours. This is an expected consequence of gradual structural degradation of the quasi-2D perovskite framework under high-intensity illumination. Furthermore, the magnetic field-tunable SF data presented in Fig. 4, which were collected within a one-hour timeframe, also attest to the high stability of our samples during measurement.

To further clarify the nature of our samples and address potential alternative explanations, we have added a new high-resolution top-down STEM image of the superlattice edge in the revised Fig. 1d. This image, to the best of our knowledge, represents the first clear STEM visualization of the edges of vertical quasi-2D perovskite structure. It distinctly shows the quasi-2D quantum well structure, conclusively ruling out the possibility that the observed edge states originate from 3D perovskite phases.

Regarding the differences between our observations and previous work, we believe these arise from variations in sample structure, composition, and measurement conditions. For instance, the prior study on edge states was based on exfoliated $BA_2FAPb_2I_7$ microplates, and the PL was measured under 400 nm UV excitation (ACS Nano 2019, 13, 2, 1635).

Finally, to ensure full transparency and reproducibility, we have updated the Methods section to include these experimental details. Unless otherwise specified (e.g., the vacuum stability test described above), all optical spectroscopy measurements in our study, including PL, TA, pump-probe, and FWM, were performed at room temperature with samples in ambient air (relative humidity ~50%).

In addition, further simulations are required to conclusively support the interpretation of the directional emission measurements. The peculiar crisscross network structure could hinder long-range propagation. In such a scenario, amplified spontaneous emission (ASE can occurs on the hundreds um-scale length) may be more efficiently scattered out of plane, potentially explaining the enhanced emission observed at normal incidence.

Response:

We agree with the reviewer that the influence of the crisscross network structure on directional emission needs to be rigorously examined. To address this, we have performed additional simulations, which are now included in the revised Supplementary Information (newly added Supplementary Fig. 13f, 13g and revised Supplementary Note 6).

Specifically, we compared the horizontal-to-vertical emission intensity ratio of our crisscross superlattice with that of a bulk crystal having the same width, length, and height. The simulations reveal no significant difference in this emission ratio between the two structures, whether the field integration is performed over the entire crystal volume or restricted to the top 50 nm. These results indicate that the crisscross network does not inherently enhance out-of-plane extraction via structural scattering. Therefore, the stronger normal-direction ASE observed in our superlattices cannot be explained by a geometry-induced scattering effect and is instead consistent with an intrinsic origin, namely macroscopic dipole formation associated with the coherent SF process.

Full simulation details, calculation procedures, and parameters are provided in the revised Supplementary Note 6.

Without addressing these important insights, the manuscript does not yet reach the quality level required for publication in Nature.

Response:

We believe we have now addressed all of the reviewer's concerns. In particular, we have:

- Shown that the theoretical formalism developed in Refs. 1,2 is applicable to our measurements by comparing multiple experimental signatures which are in excellent quantitative agreement with those reports.
- Provided additional data and experimental details regarding sample preparation to ensure reproducibility.
- Performed the requested simulations to definitively rule out ASE.

Furthermore, we wish to emphasize that the central advance of our manuscript is the first experimental realization of chiral superfluorescence in any system—at room temperature or otherwise. Conventional room-temperature SF in hybrid perovskites, along with the associated long coherence times, has already been demonstrated in Refs. 1–2 and given a plausible theoretical explanation. We now also provide an explanation for the role of edge states in enabling longer coherence times. Our work builds upon this foundation by introducing chirality as a new degree of control over the cooperative emission process.

We thank the reviewer once more for their constructive comments, which have significantly improved the manuscript.

Referee #2 (Remarks to the Author):

I appreciate the authors' meticulous and comprehensive response to my previous report. In the revised manuscript and supplementary materials, they have significantly strengthened the analysis and addressed the key concerns I raised regarding the identification and interpretation of superfluorescence.

The revision now presents multiple, consistent lines of evidence supporting the attribution of the observed emission to superfluorescence rather than to coherent artifacts, amplified spontaneous emission, or photonic effects. These include fluence-dependent scaling and delay-time behavior aligned with SF theory, clear experimental differentiation from ASE through directionality measurements and controls, time-resolved signatures of collective emission, and independent coherence measurements confirming that the relevant timescales for SF are met.

The authors have also clarified the role of sample geometry and ruled out trivial photonic or waveguiding origins of the observed polarization and directionality. While some aspects of the chiral, forward-directed emission are discussed at a qualitative level, they are clearly framed as interpretations and do not affect the central identification of superfluorescence.

Overall, the manuscript is now internally consistent, well supported experimentally, and significantly improved. I believe the authors have adequately addressed my earlier concerns and support publication in its present form.

Response:

We thank the referee for their thoughtful assessment and for recommending publication.

Referee #3 (Remarks to the Author):

Authors have performed multiple new experiments as well as theoretical modeling to address the comments of all the reviewer. These new experiments and theory work address all the key concerns of the reviewer and substantially improved the paper. Now I believe that the paper provide not only a strong evidence of chiral SF but also a plausible theoretical explanation. This work represents a significant advance of the field worthy of publication in nature. I can now recommend the paper as it is.

Response:

We thank the reviewer for their thoughtful feedback and strong support for publication.

Referee #1 (Remarks to the Author):

The authors have clarified their views on long exciton coherence times. Without further technical comments, I would support publishing this version.

Response:

We thank Referee #1 for acknowledging our revisions, for supporting publication, and for their constructive feedback, which has significantly strengthened the paper.

Referee #2 (Remarks to the Author):

The authors have revised the manuscript and have addressed the concerns raised by Referee #1. The coherence properties, including the dephasing time, are sufficient to support the observation of superfluorescence, as the condition that the coherence time exceeds the buildup time is satisfied. The addition of four-wave mixing measurements provides a direct and independent confirmation, further strengthening the experimental basis. Together with the threshold behavior, superlinear scaling, delay dynamics, and directionality, the identification of superfluorescence is well supported.

The main advance of the work, namely, the demonstration of chiral superfluorescence and its control via material handedness and magnetic field, is clearly established. The emergence of circular polarization, its sign reversal, and its tunability are unambiguous and represent a significant result, supported by a consistent theoretical description.

The question regarding the origin of the long coherence time pertains to a deeper microscopic understanding rather than the validity of the central claim. The authors provide a physically reasonable explanation grounded in prior literature, which is sufficient for the scope of the present work. In my view, the points raised by Referee #1 have been adequately addressed.

Response:

We thank Referee #2 for their careful evaluation and positive assessment.