

Resolving intrinsic dislocation structure in perovskite crystals using pulsed electron beam with atomic resolution

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Attachments originally included by the reviewers as part of their assessment can be found at the end of this file.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

dislocation structure in perovskite materials through pulsed electron beam technology implemented by electrostatic dose modulator (EDM), and revealed the slip climb transition mechanism induced by continuous electron beam, which has important scientific significance and technical value. I will recommend this work after the following comments are fully addressed:

1. In my understanding, EDM controls the electron beam through electrostatic deflection. What is the basis for selecting the core parameters of EDM, such as a 25% duty cycle for CsPbBr₃ and a 20% duty cycle for SrTiO₃? Please provide explanations of the relevant mechanisms.
2. When comparing pulsed beams (e.g., $1.1 \times 10^{10} \text{ e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1}$ in the CsPbBr₃ experiment) with continuous beams (e.g., $4.5 \times 10^{11} \text{ e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1}$), the dose rate difference is 41 times greater. Does this difference introduce additional factors beyond point defect concentration? Has the effect of total electron dose rather than dose rate on dislocation structures been excluded through controlled experiments with a consistent total dose?
3. The experiment observed a significant difference in the structure of dislocations terminated by anions and cations after climb (compact vs. extended), but the explanation of the underlying physical mechanism is relatively brief. Is this structural difference related to the charge state, local bonding strength, or point defect segregation behavior? They should comment regarding this in the manuscript.
4. In SrTiO₃, the authors observed a near glide-dissociated configurations and attributed it to its higher beam sensitivity. What are the specific structural differences between the near glide and ideal glide? Is there a quantitative standard to distinguish? Why do higher defect generation rates and lower recombination rates only result in a slight deviation from collinearity, rather than directly triggering a climb transition? Furthermore, is there a quantitative relationship between electron dose/dose rate or point defect concentration and the density of the two types of dislocations?
5. The article suggests that EDM could be applicable to other beam-sensitive materials, such as Al₂O₃. However, given that perovskites have soft lattices and Al₂O₃ is a high-hardness ceramic with markedly different atomic displacement cross-sections and defect diffusion kinetics, it would be essential to optimize pulse parameters specifically for each material. What strategy should be adopted to optimize parameters such as the duty cycle and the beam-off interval for such materials? Furthermore, are there universal criteria for designing these parameters across different material systems?
6. Lead halide perovskites are typical beam-sensitive materials. Recently, atomic imaging of halide perovskites has been widely reported by using low-dose methods, such as iDPC-STEM. These works should be well introduced. Please conduct a comprehensive literature search and citation as much as possible.

Reviewer #2

(Remarks to the Author)

The paper describes interesting in-situ STEM studies that explore the impact of the chemistry of dislocation core its motion. They show that continuous beams instead triggers rapid glide-to-climb transitions, where the climbed structures depend on core chemistry: anion-terminated dislocations

remain compact, while cation-terminated ones extend. These findings elucidate the intrinsic dislocation structures and deformation mechanisms in perovskites. Their work is supported by atomistic simulation studies.

There are however some issues that need clarification:

1. while it is understood that concerns about radiation damage provides limits, there is no discussion on the use of diffraction contrast imaging to provide direct evidence of dislocation burger's vectors. The experimental part of the paper provides no discussion of partial, domain wall interactions which are known to be important to contribute to deformation mechanisms in perovskites structure
2. Given that the experiments were conducted on single crystal nanopillars, no STEM experimental characterization on configurations and reactions associated with activity related to interactions of non-coplanar dislocations
3. A discussion on the nature of primary and secondary slip planes is missing and linking schmid factor analysis would make the experimental work far more compelling

Reviewer #3

(Remarks to the Author)

See attached.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The author's response has addressed the questions that I raised in the last version, and I believe it is recommended to publish this work in NC.

Reviewer #3

(Remarks to the Author)

See attached

Reviewer #4

(Remarks to the Author)

The manuscript presents an in-situ STEM investigation of dislocation behavior in perovskite materials using pulsed and continuous electron beam irradiation controlled via an electrostatic dose modulator (EDM). The authors demonstrate that pulsed electron beams stabilize glide-dissociated dislocations, whereas continuous irradiation induces a glide-to-climb transition. Importantly, the study reveals that the climbing mechanism and resulting dislocation structures are strongly governed by dislocation core chemistry, with anion-terminated cores remaining compact and cation-terminated ones exhibiting extended configurations. Supported by atomistic simulations, these findings provide new insight into intrinsic dislocation structures and deformation mechanisms in perovskites, highlighting both fundamental scientific relevance and technical significance. The authors have satisfactorily addressed all concerns raised by the referees and have substantially strengthened the manuscript through additional measurements and calculations. In its current form, the work represents a significant and high-impact contribution to the field and is suitable for publication in Nature Communications.

Version 2:

Reviewer comments:

Reviewer #3

(Remarks to the Author)

The authors have resolved all my comments with the manuscript, and I can now recommend it for publication.

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Responses to Reviewers' Comments (NCOMMS-25-85515-T)

Below are our point-by-point responses (shown in **black** text) to the reviewers' comments (shown in **blue** text).

Reviewer #1 (Remarks to the Author):

Dislocation structure in perovskite materials through pulsed electron beam technology implemented by electrostatic dose modulator (EDM), and revealed the slip climb transition mechanism induced by continuous electron beam, which has important scientific significance and technical value. I will recommend this work after the following comments are fully addressed.

Response:

We sincerely thank the reviewer for the positive feedback and insightful suggestions. We particularly appreciate the valuable comments regarding the need to better control variables and the physical origin of distinct dissociation widths in climb-dissociated dislocations, which have significantly strengthened the methodological rigor of the manuscript and in-depth discussion of the results.

1. In my understanding, EDM controls the electron beam through electrostatic deflection. What is the basis for selecting the core parameters of EDM, such as a 25% duty cycle for CsPbBr₃ and a 20% duty cycle for SrTiO₃? Please provide explanations of the relevant mechanisms.

Response:

We appreciate the reviewer's insightful question regarding the basis for selecting electrostatic dose modulator (EDM) parameters. Indeed, the EDM functions as a fast beam blanking system utilizing a pre-sample electrostatic deflector, which chops the continuous electron beam into pulses, thereby effectively reducing the overall electron dose rate while maintaining the ability to achieve high-resolution imaging.

The primary criterion for selecting EDM core parameters, such as duty cycle, is to resolve the intrinsic dislocation structures at atomic resolution without altering it. This involves a trade-off: on one hand, visualizing the "intrinsic dislocation structure" requires minimizing the dose rate to reduce point defect generation and preserve the pristine state. Lower EDM duty cycles and lower imaging magnifications are preferred to decrease the electron dose rate; on the other hand, achieving "atomic resolution" demands a sufficiently high electron dose rates, for which, higher duty cycles and magnifications are needed to enhance signal-to-noise ratio (SNR) and clearly resolve individual atoms.

For SrTiO₃, as detailed in the line 259-271 in manuscript, the material exhibits high sensitivity to electron irradiation due to the elevated point defect generation rate (K_0)

for oxygen atoms. Extensive trials were conducted to identify the minimal dose rate conditions capable of resolving the pristine glide-dissociated dislocations while maintaining lattice visibility. A 20% EDM duty cycle combined with 3M magnification emerged as the lowest-dose-rate condition that can capture the near glide-dissociate dislocation structure, meanwhile still can resolve crystal lattices. Higher EDM duty cycle or higher magnification will trigger fast dislocation climb, and lower duty cycle and magnification failed in resolving crystal lattices. In contrast, CsPbBr₃ demonstrates greater beam stability owing to its relatively lower K_0 for bromine atoms, providing a wider safety margin against damage. Consequently, a slightly higher EDM duty cycle of 25% and 5M magnification were employed in the dataset in original Fig. 2 to improve image SNR, enabling clearer atomic visualization without altering the intrinsic dislocation structures.

2. When comparing pulsed beams (e.g., $1.1 \times 10^{10} \text{ e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1}$ in the CsPbBr₃ experiment) with continuous beams (e.g., $4.5 \times 10^{11} \text{ e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1}$), the dose rate difference is 41 times greater. Does this difference introduce additional factors beyond point defect concentration? Has the effect of total electron dose rather than dose rate on dislocation structures been excluded through controlled experiments with a consistent total dose?

Response:

We thank the reviewer for raising this important comment regarding the dose rate difference ($\sim 41\times$ higher in the continuous beam case) and its potential confounding effects beyond point defect concentration, as well as the role of total accumulated electron dose versus dose rate.

We acknowledge and apologize that our initial comparison inadvertently included uncontrolled variables, as also indicated by Reviewer #3: the continuous beam images were acquired with higher magnification and with a larger image size (2048×2048 pixels) compared to the pulsed beam case (1024×1024 pixels), resulting in a substantially higher dose rate difference of ~ 41 times. This difference primarily led to a stronger point defect generation rate, which has already been quantitatively accounted for in our defect kinetics calculations (Supplementary Note 5). From our experimental observations, this was the only detectable consequence: an elevated point defect concentration that drove instant glide-to-climb transition. No other distinct influences, such as significant charging effects, or structural decomposition were observed under these conditions.

To rigorously isolate this effect of extra high dose rate, we have conducted new controlled experiments where all parameters were kept identical (same magnification, image size of 1024×1024 pixels, pixel dwell time of $4 \mu\text{s}$), varying only the EDM on/off status. We configured the EDM pulse period as $4 \mu\text{s}$ to synchronize it with the pixel dwell time, for the purpose of pixel-by-pixel dose rate modulation. The EDM duty cycle is 25%, namely, $1 \mu\text{s}$ beam-on followed by $3 \mu\text{s}$ beam-off during each pulse

period/pixel dwell time. Under these settings, the continuous beam dose rate is exactly $4\times$ higher ($1.14 \times 10^{11} \text{ e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1}$) than that of pulsed beam ($2.86 \times 10^{10} \text{ e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1}$). As shown in Figs. R1a-b, under pulsed beam conditions, the intrinsic glide-dissociated dislocation structure was stably captured throughout 26 sequential frames. Upon turning off the EDM and switching to continuous beam, the dislocation underwent a gradual glide-to-climb transition during subsequent imaging (Figs. R1c-d).

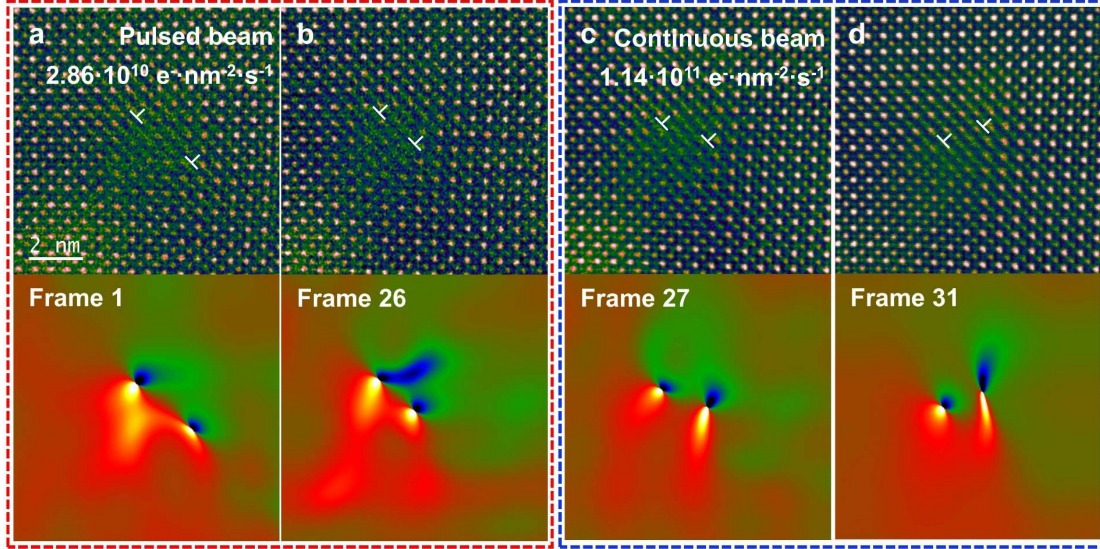


Fig. R1. New controlled experiments in CsPbBr₃, with identical imaging parameters for pulsed and continuous beam except EDM on/off. **a**, The first frame under pulsed beam condition ($4 \mu\text{s}$ period, 25% duty cycle, dose rate $2.86 \times 10^{10} \text{ e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1}$). **b**, The 26th frame under pulsed beam condition. **c-d**, Upon switching to continuous beam (EDM off; dose rate $1.14 \times 10^{11} \text{ e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1}$), the same dislocation underwent gradual glide-to-climb transition.

To fully deconvolve the effects of total electron dose, dose rate, and beam pulsing on dislocation motion as also suggested by editor, we conducted new experiments and compared their results across datasets with different beam conditions. All crucial beam parameters are listed in Table R1, i.e., the beam type, dose rate, beam-on time within a dwell time, and total dose accumulated on each pixel. t_0 refers to the time to reach “quasi-steady-state”, at which point defect concentration reached peak value of C_0 (details in Supplementary Note 5). And Δt_s stands for the sustained duration of peak value C_0 within a pixel dwell time.

- **Dataset 1:** pulsed beam data in original Fig. 2. Total dose on each pixel after 21 frames was $\sim 2.33 \times 10^5 \text{ e}^- \cdot \text{nm}^{-2}$ ($1.11 \times 10^{10} \text{ e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1} \times 1 \mu\text{s} \times 21$) with the glide-dissociated structure fully preserved.
- **Dataset 2 (1):** newly collected pulsed beam data (Figs. R1a-b). The glide-dissociation remained stable throughout 26 frames, accumulating a total dose of $\sim 7.44 \times 10^5 \text{ e}^- \cdot \text{nm}^{-2}$ per pixel.
- **Dataset 2 (2):** when EDM was off and the pulsed beam turned into continuous beam, glide-to-climb started (Figs. R1c-d). The total dose after one frame under the continuous beam is $\sim 4.56 \times 10^5 \text{ e}^- \cdot \text{nm}^{-2}$ ($1.14 \times 10^{11} \text{ e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1} \times 4 \mu\text{s} \times 1$).

- **Dataset 3:** newly collected continuous beam data shown in Fig. R2a, with the same rate of Dataset 2(1). Climb dissociation occurred at frame 1, accumulating total dose of only $\sim 1.14 \times 10^5 \text{ e}^- \cdot \text{nm}^{-2}$ per pixel.
- **Dataset 4:** newly collected continuous beam data shown in Fig. R2b, with the dose rate of four times of Dataset 1. All imaging setups are the same as Dataset 1 except EDM was turned off. Climb dissociation occurred at frame 1, with a cumulative total dose of only $\sim 1.77 \times 10^5 \text{ e}^- \cdot \text{nm}^{-2}$ per pixel.

Table R1 Dislocation structure and point defect generation under various beam conditions

Dataset	Beam	Dose rate ($\text{e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1}$)	Beam-on time (μs)	Total dose ($\text{e}^- \cdot \text{nm}^{-2}$)	t_0 (μs)	Δt_s (μs)	C_0 (10^{-5})	Structure
1	Pulsed	$1.11 \cdot 10^{10}$	1	$2.33 \cdot 10^5$	0.73	0.27	1.54	glide
2 (1)	Pulsed	$2.86 \cdot 10^{10}$	1	$7.44 \cdot 10^5$	0.46	0.54	2.47	glide
3	Continuous	$2.86 \cdot 10^{10}$	4	$1.14 \cdot 10^5$	0.46	3.54	2.47	climb
4	Continuous	$4.43 \cdot 10^{10}$	4	$1.77 \cdot 10^5$	0.37	3.63	3.08	climb
2 (2)	Continuous	$1.14 \cdot 10^{11}$	4	$4.56 \cdot 10^5$	0.23	3.77	4.95	climb

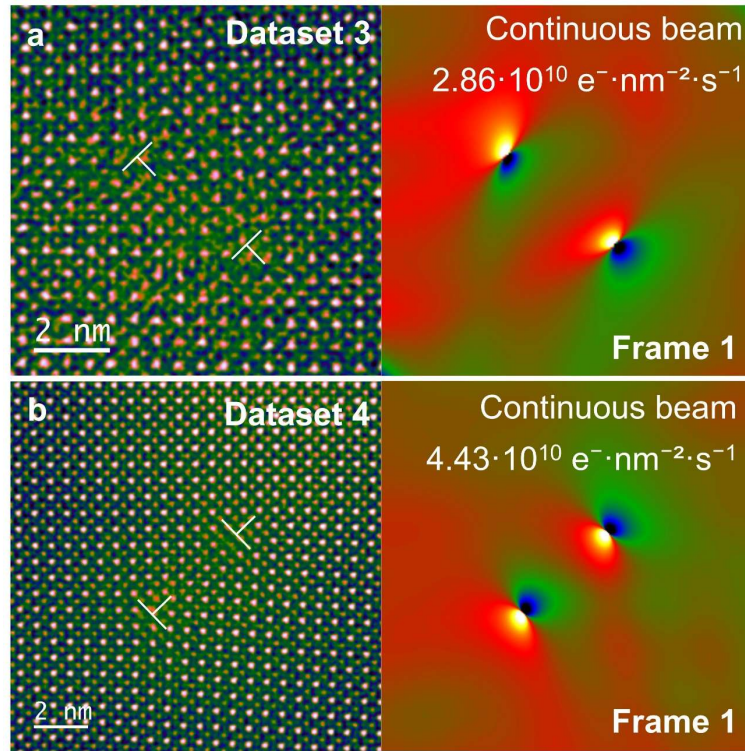


Fig. R2. New collected experiments in CsPbBr_3 under continuous beam. **a**, Dislocation structure under electron dose rate of $2.86 \times 10^{10} \text{ e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1}$, which climbed on first frame. **b**, Dislocation structure under electron dose rate of $4.43 \times 10^{10} \text{ e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1}$, which climbed on first frame.

Firstly, we can exclude total dose as the cause of dislocation climb. To support this, Dataset 1 accumulated a total dose of $2.33 \cdot 10^5 \times 10^{10} \text{ e}^- \cdot \text{nm}^{-2}$ after 21 frames under pulsed beam, with the glide structure preserved (bottom blue box in Fig. R3). In Dataset 2(1), the same total dose is reached at the 8th frame (middle red box Fig. R3), and upon

further imaging, the dislocation maintained its intrinsic structure up to 26 frames, accumulating $7.44 \cdot 10^5 \times 10^{10}$ (top red box in Fig. R3). However, in Datasets 2(2), 3, and 4 under continuous beam with high dose rate, climb occurred instantly, even though the total dose was far below that accumulated under pulsed beam. These results indicate a threshold dose rate: below it, dislocations can be imaged safely regardless of total dose; above it, climb occurs.

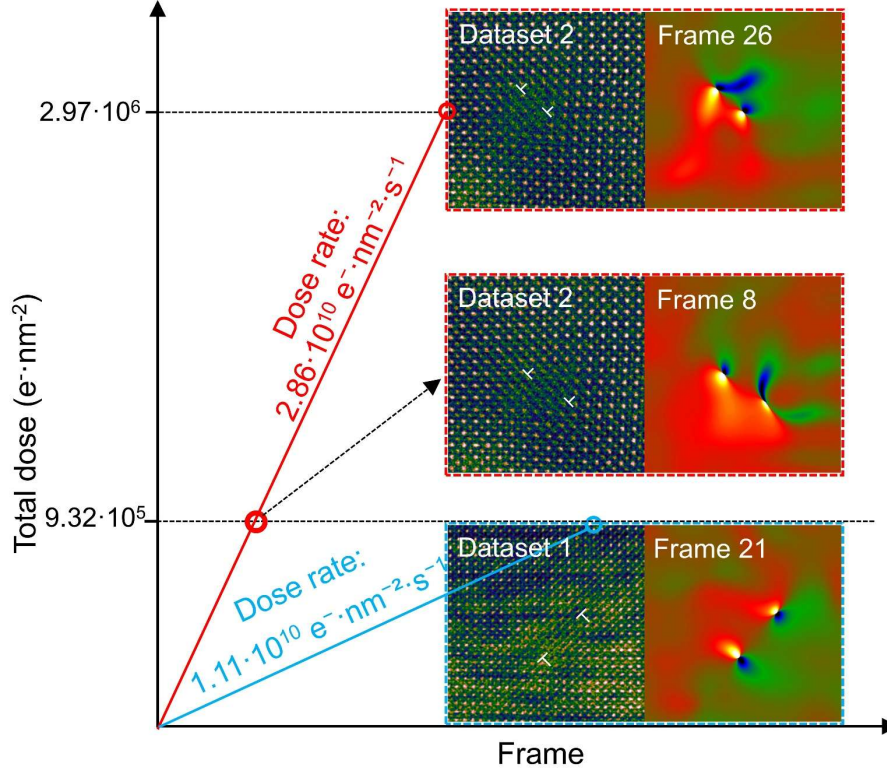


Fig. R3. Dislocation structures under pulsed beam conditions at different cumulative total doses. The bottom image in blue box is the 21st frame from Dataset 1. The middle and top images (red box) are the 8th and 26th frames from Dataset 2(1), respectively. These demonstrate that the glide-dissociated structure remains stable even as total dose accumulates significantly beyond levels at which climb occurs under continuous beam.

Second, climb requires not only defect generation but also sufficient time for defects to diffuse to the dislocation core, accumulate to supersaturation and drive non-conservative climb motion. Therefore, beam-on time per pixel, or the sustained duration (Δt_s) of peak defect concentration, is a critical parameter beyond just dose rate. As shown in Table R1 and Fig. R4, Datasets 2(1) and 3 have the same dose rate, t_0 and C_0 , yet climb occurred only under continuous beam (Dataset 3), not pulsed beam (Dataset 2 (1)). This difference is due to Δt_s . Within a 4 μs dwell time, the peak defect concentration C_0 under pulsed beam sustained only $\Delta t_s = 0.54 \mu s$, the beam-off interval enables rapid point defect recombination before climb. In contrast, the peak defect concentration C_0 sustained $\Delta t_s = 3.54 \mu s$ under continuous beam, providing the time needed for defects to migrate to the core, exceed the critical supersaturation threshold, and activate climb.

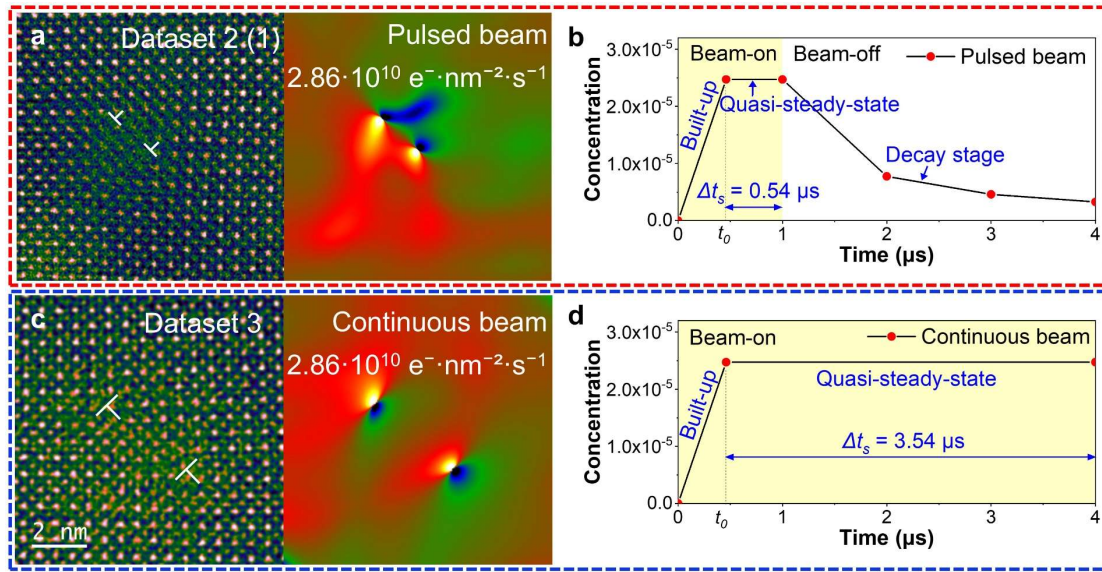


Fig. R4. Dislocation structures under pulsed beam and continuous conditions at the same electron dose rate of $2.86 \times 10^{10} \text{ e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1}$. **a**, The 26th frame from Dataset 2(1) under pulsed beam (4 μs period, 25% duty cycle), showing the glide-dissociated structure remains stable. **b**, Point defect concentration versus time within one 4 μs pixel dwell time for Dataset 2(1), with peak concentration sustained for $\Delta t_s \approx 0.54 \mu\text{s}$. **c**, The 1st frame from Dataset 3 under continuous beam, climb-dissociation has already occurred. **d**, Point defect concentration versus time within one 4 μs pixel dwell time for Dataset 3, peak concentration sustained for $\Delta t_s \approx 3.54 \mu\text{s}$.

From the comparison of results, we can clearly draw a conclusion that pulsing nature, which reduces the dose rate and provides beam-off intervals for defect annihilation, is the primary factor suppressing climb, rather than simply dose rate or total dose. With these new controlled results, we are able to deconvolve the effect of total dose and dose rate on dislocation behaviour. We have incorporated this thorough discussion and newly collected results as Supplementary Note 6, and added a brief discussion in line 246-257 of revised manuscript. The well-controlled Dataset 2 were used as Fig. 2. The dataset in original Fig. 2 was relocated to Fig. R4, though it failed to strictly control variables, this dataset does perfectly uncover the intrinsic dislocation structure and revealed beam-induced climb behaviour. Thank you for this constructive comment, which has greatly strengthened the rigor of this manuscript.

3. The experiment observed a significant difference in the structure of dislocations terminated by anions and cations after climb (compact vs. extended), but the explanation of the underlying physical mechanism is relatively brief. Is this structural difference related to the charge state, local bonding strength, or point defect segregation behavior? They should comment regarding this in the manuscript.

Response:

We thank the reviewer for this constructive comment, which facilitated a more detailed explanation of the physical mechanism underlying the distinct climb-dissociated configurations observed for anion-terminated (compact) and cation-terminated

(extended) dislocations. To address this, we performed charge density analyses on climb-dissociated dislocation models in SrTiO₃. These calculations reveal that the structural differences arise primarily from electrostatic interactions governed by the charge states of the dislocation cores and the associated antiphase boundary (APB). Specifically, we projected the nominal ionic charges of all atoms onto the (001) plane and convoluted them with 2-D Gaussian kernels ($\sigma = 3 \text{ \AA}$) to obtain a smooth charge density map $\Delta\rho(x,y)$:

$$\Delta\rho(x,y) = \sum_{i=1}^N \frac{q_i}{\sigma\sqrt{2\pi}} \exp\left(-\frac{(x_i - x)^2 + (y_i - y)^2}{2\sigma^2}\right)$$

where q_i is the formal charge of atom i . The kernel width is chosen so that $\Delta\rho$ vanishes in bulk regions.

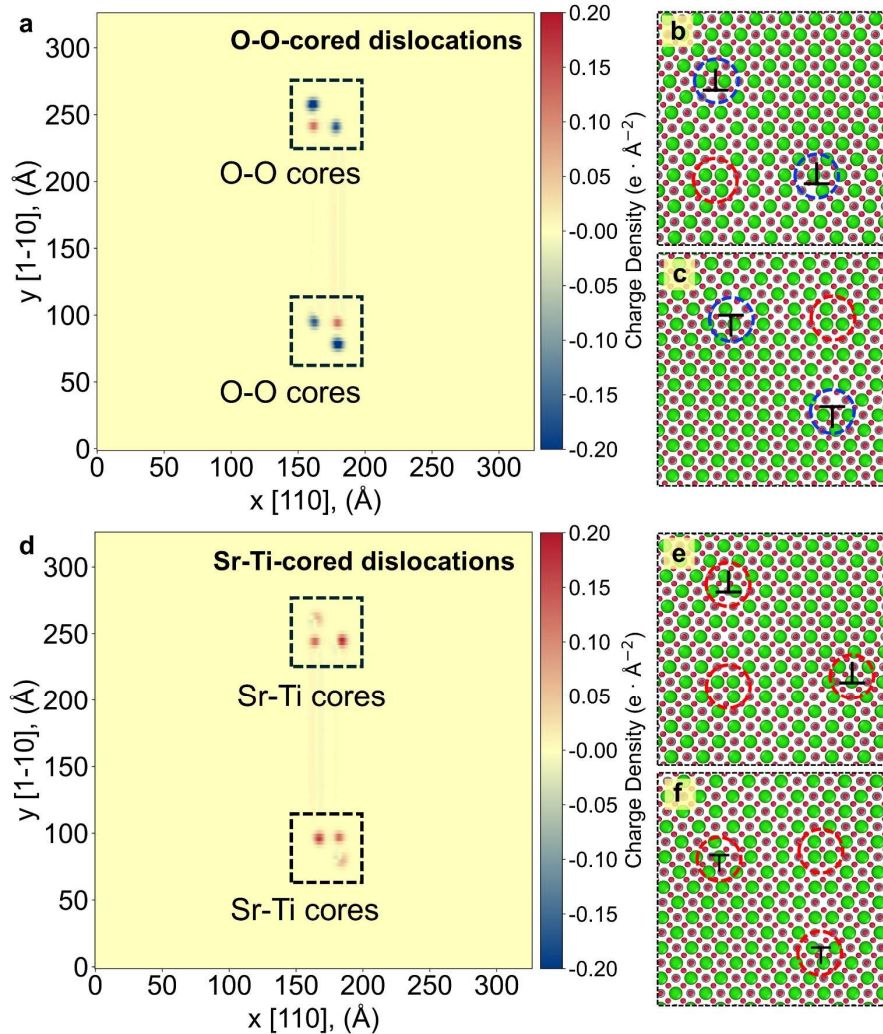


Fig. R5 Charge density analyses of distinct climb-dissociated dislocations in SrTiO₃. **a**, Charge density map of a model containing two climb-dissociated O-O-cored dislocations. **b-c**, Magnified atomic arrangements around the two dislocations in dashed squares in **(a)**. **d**, Charge density map of a model containing two climbed Sr-Ti-cored dislocations. **e-f**, Magnified atomic arrangement around the two dislocations in dashed squares in **(d)**.

For O-O-cored climb-dissociated dislocations, the charge densities before energy minimization are shown in Fig. R5a, with the atomic arrangement around the two dislocations in dashed squares are magnified in Figs. R5b-c. The partial dislocations carry localized negative charges, while the APB junction introduces a region of positive charge. The opposite charge signs result in attractive Coulombic interactions that will pull the partials together, stabilizing a compact configuration, consistent with our experimental observations (Figs. 4a-e) and MD energy profiles (Fig. 4m). In contrast, Sr-Ti-cored dislocations distinctly exhibit three positive charge concentrations: two from the partial cores themselves and one from the APB junction (Figs. R5d-f). The resulting like-charge Coulombic repulsion pushes the partials apart, leading to a wider, extended configuration. Our analysis indicates that core charge states and the ensuing electrostatics dominate the equilibrium dissociation width.

We have included these findings and expanded the discussion in the revised manuscript (line 318-325 and line 334-338, Figs. 4k-l, Figs. 4n-o) and Supplementary Information (Supplementary Note 9). This addition promoted a more profound mechanistic interpretation and significantly strengthened the paper.

4. In SrTiO₃, the authors observed a near glide-dissociated configurations and attributed it to its higher beam sensitivity. What are the specific structural differences between the near glide and ideal glide? Is there a quantitative standard to distinguish? Why do higher defect generation rates and lower recombination rates only result in a slight deviation from collinearity, rather than directly triggering a climb transition? Furthermore, is there a quantitative relationship between electron dose/dose rate or point defect concentration and the density of the two types of dislocations?

Response:

The ideal glide-dissociated configuration for a $\{110\}\langle\bar{1}\bar{1}0\rangle$ dislocation consists of two partial dislocations that are strictly collinear and parallel along the slip direction on the glide plane (Fig. 3e). The “near glide” morphology observed in Fig. 3f features a slight deviation from collinearity, representing a transitional state where climb has already begun, resulting in subtle non-parallel alignment. There is no strict quantitative standard (e.g., fixed angular deviation or separation distance) to distinguish near-glide from ideal glide, as the transition is continuous until reaching a steady climb state. Instead, the distinction is primarily qualitative, based on morphological features in STEM-HAADF images: ideal glide exhibits perfect parallelism and collinearity, while intermediates with minor deviations indicate ongoing transition.

To address why higher defect generation and lower recombination rates cause only slight deviation rather than full climb, note that climb velocity is proportional to dose rate above a critical threshold. For SrTiO₃, even our minimum lattice-resolvable dose rate used in Fig. 3f still exceeds the threshold. However, the pulsed beam slows climb kinetics enough to capture the intermediate near-glide state, providing key evidence of

the intrinsic structure. We have clarified in the revised manuscript that the dislocation in Fig. 3f is already undergoing incipient climb under these conditions, see line 285-288, and line 296-299 in revised manuscript.

For CsPbBr₃, the quantitative relationship between dose/dose rate, point defect concentration, and dislocation type prevalence can be inferred from Table R1. A dose rate of $2.86 \times 10^{10} \text{ e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1}$ at 25% pulsed beam duty cycle appears to be a safe threshold for imaging intrinsic structures; exceeding it might trigger climb. In SrTiO₃, however, optimized EDM pulsed beam conditions still exceed this inferred threshold, yielding partial climb and near-glide forms. Future low-dose advancements (e.g., enhanced detectors or reduced currents) may allow precise threshold quantification. We have added this discussion to the revised manuscript for clarity.

5. The article suggests that EDM could be applicable to other beam-sensitive materials, such as Al₂O₃. However, given that perovskites have soft lattices and Al₂O₃ is a high-hardness ceramic with markedly different atomic displacement cross-sections and defect diffusion kinetics, it would be essential to optimize pulse parameters specifically for each material. What strategy should be adopted to optimize parameters such as the duty cycle and the beam-off interval for such materials? Furthermore, are there universal criteria for designing these parameters across different material systems?

Response:

We thank the reviewer for the insightful comments regarding the applicability of the electrostatic dose modulator (EDM) pulsed-beam technique to other beam-sensitive materials, such as Al₂O₃, and for highlighting the need to account for differences in lattice softness, atomic displacement cross-sections, and defect diffusion kinetics. We fully agree that pulse parameters (e.g., duty cycle and beam-off interval) must be optimized specifically for each material, as these factors govern the balance between minimizing beam-induced defect accumulation (to preserve intrinsic dislocation structures) and maintaining sufficient dose rates for adequate signal-to-noise ratios in imaging. As detailed in the main text and Supplementary Notes 4 and 5, our analytical framework, which is based on analytical calculations of atomic displacement cross sections and point-defect generation/recombination kinetics, can provide a robust, material-specific strategy for this optimization. These calculations incorporate crystallographic structure and bonding characteristics, enabling quasi-quantitative predictions of beam sensitivity and defect dynamics.

Firstly, the atomic displacement cross section takes bonding characteristics and crystal lattice arrangements into consideration, and exactly determines the probability and dominant species of point defects generated under electron irradiation for each material. For instance, lighter elements are generally more easily displaced, but in Al₂O₃, Al atoms (despite being heavier) have lower displacement energy E_d of 18 eV than O atoms ($\sim 76 \text{ eV}$)^{1,2}, due to their occupancy of relatively open octahedral sites and lower-energy interstitial paths. Table R2 illustrates the atomic displacement cross

section of Al and O in Al₂O₃, using the calculation process illustrated in Supplementary Note 4. From the table, it can be seen that the Al point defect generated by 300 KeV incident electron beam dominates dislocation climb with cross section of 20 barn, and O is unlikely or very slightly to be displaced with -1.4 barn. This contrasts with perovskite oxides like SrTiO₃, in which oxygen point defects typically dominate dislocation climb.

Table R2 Atomic displacement cross sections for Al₂O₃ under a 300 KeV electron beam

	E_d (eV)	E_{max} (eV)	E_t (KeV)	σ (barn)
Al	18	31.54	187	20.0
O	76	53.23	398	-1.4

Secondly, defect kinetics further inform parameter selection: the generation rate (k_0), quasi-steady-state buildup time (t_0), migration energies (E_{mi}), and interstitial-vacancy recombination coefficients (K_{iv}) predict whether defects accumulate and concentration evolution during the beam-on period or relax during beam-off intervals. To illustrate, here we compare the defect kinetics using the EDM parameters from our SrTiO₃ experiments in Figure 3 (4 μ s pulse period, 20% duty cycle with 0.8 μ s beam on and 3.2 μ s beam off, electron dose rate $\Phi = 1.3 \cdot 10^{10} \text{ e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1}$). As shown in Table R3, the oxygen defect generation rate in SrTiO₃ is ~ 36.3 dpa/s (with $t_0 \approx 2.1 \mu$ s), while the Al defect rate in Al₂O₃ is only ~ 8.5 dpa/s ($t_0 \approx 2.4 \times 10^7 \mu$ s, due to much higher migration energy E_{mi} of 1.1 eV³ and lower recombination coefficient K_{iv}). During the 0.8 μ s beam-on time, defect concentrations remain in the build-up stage for both materials, but Al₂O₃ proves more robust with a much lower defect concentration (defects per atom) of $6.8 \cdot 10^{-6}$, than SrTiO₃ ($2.9 \cdot 10^{-5}$). Thus, for Al₂O₃, EDM duty cycles could be safely increased, for example to 25–40%, to yield higher average dose rates and improved image quality while suppressing unwanted climb dissociation.

Regarding universal criteria: while parameters must ultimately be tailored experimentally to each material, the cross section and kinetics calculations serve as broadly applicable design principles. They encapsulate material-specific properties (e.g., displacement thresholds, migration barriers) and can guide initial parameter selection across systems, including perovskites and harder ceramics like Al₂O₃. This framework makes EDM a versatile tool for beam-sensitive materials and has clarified its general applicability in the revised manuscript.

Table R3 Point defect concentration dynamics in SrTiO₃ and Al₂O₃

	Defect species	E_{mi} (eV)	D_i (nm ² /s)	K_{iv} (s ⁻¹)	K_0 (dpa/s)	t_0 (μ s)	C (at 0.8 μ s)
SrTiO ₃	O	0.3	$1.5 \cdot 10^7$	$6 \cdot 10^9$	36.3	2.1	$2.9 \cdot 10^{-5}$
Al ₂ O ₃	Al	1.1	$2.9 \cdot 10^{-7}$	$2.0 \cdot 10^{-4}$	8.5	$2.4 \cdot 10^7$	$6.8 \cdot 10^{-6}$

6. Lead halide perovskites are typical beam-sensitive materials. Recently, atomic imaging of halide perovskites has been widely reported by using low-dose methods,

such as iDPC-STEM. These works should be well introduced. Please conduct a comprehensive literature search and citation as much as possible.

Response:

Thank you for helping us refine the literature background. Following this suggestion, we have conducted a literature review and incorporated several relevant studies on atomic-resolution structural characterization with iDPC-STEM technique of CsPbBr₃ and CsPbI₃ halide perovskites. These advancements have enabled detailed structural characterizations, including phase transition under heating, and precipitation behaviors and phase/crystal interfaces⁴⁻⁷. We also add the following discussion into introduction in revised manuscript, as highlighted from line 81 to line 84:

“Recent low-dose techniques, notably integrated differential phase contrast (iDPC)-STEM, have proven particularly effective for visualizing light elements and lattice structures with improved signal-to-noise ratios and reduced overall damage in halide perovskites²³⁻²⁶.”

Reviewer #2 (Remarks to the Author):

The paper describes interesting in-situ STEM studies that explore the impact of the chemistry of dislocation core its motion. They show that continuous beams instead triggers rapid glide-to-climb transitions, where the climbed structures depend on core chemistry: anion-terminated dislocations remain compact, while cation-terminated ones extend. These findings elucidate the intrinsic dislocation structures and deformation mechanisms in perovskites. Their work is supported by atomistic simulation studies.

There are however some issues that need clarification:

1. While it is understood that concerns about radiation damage provides limits, there is no discussion on the use of diffraction contrast imaging to provide direct evidence of dislocation burger's vectors. The experimental part of the paper provides no discussion of partial, domain wall interactions which are known to be important to contribute to deformation mechanisms in perovskites structure

Response:

We thank the reviewer for raising this point about diffraction contrast imaging. Diffraction contrast imaging, utilizing weak-beam or two-beam techniques, is indeed a powerful method for statistical analysis of dislocation characters and Burgers vector determination, particularly in plastically deformed bulk materials where high dislocation densities accumulate.

In metallic materials with non-directional metallic bonds, multiple independent slip systems exist. The most close-packed plane is preferred. However, perovskites are typically ionic crystals, where the {011}<0 $\bar{1}$ 1> system is universally the primary slip

system for room-temperature deformation. Although not the shortest lattice translation, this system is the only one that prevents close approach of like-charged cations during glide, thus introducing minimum Columb repulsion⁸. As early studies have noted, “There is now ample evidence showing that all crystals with perovskite structure slip on the $\{011\}\langle 0\bar{1}1\rangle$ system at room temperature, irrespective of whether they are ideal or distorted with the BaTiO₃ or GdFeO₃ distortion”⁹, as verified by weak-beam bright-field (BF) TEM diffraction contrast in these studies.

Recent atomic-resolution STEM-HAADF observations of room-temperature deformed perovskites further support the $\{011\}\langle 0\bar{1}1\rangle$ slip system¹⁰⁻¹⁴. In this study, we employed atomic-resolution STEM to provide direct real-space imaging of the dislocation core, enabling precise Burgers vector determination. As shown in Fig. R6, the atomic columns around the core are clearly resolved. The yellow Burgers circuit demonstrates closure failure and confirms the vector $\mathbf{b} = \mathbf{a}\cdot\langle 011\rangle$, (where $\mathbf{a}$ is the lattice constant). The cyan Burgers circuits further identify the dissociation of full dislocation into two partials with $\mathbf{b} = \frac{1}{2} \mathbf{a}\cdot\langle 011\rangle$. This approach yields unambiguous evidence for Burgers vector analysis.

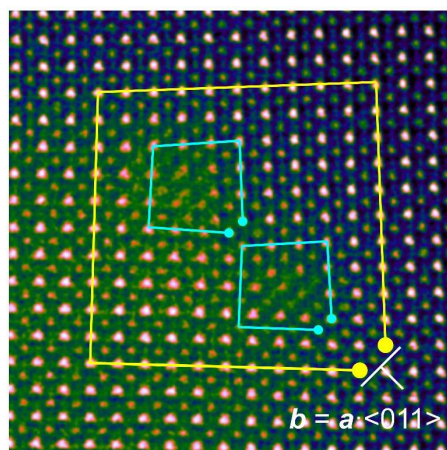


Fig. R6. Burgers vector determination of dislocation in CsPbBr₃. The yellow Burgers circuit demonstrates a closure failure corresponding to the full dislocation with Burgers vector $\mathbf{b} = \mathbf{a}\cdot\langle 011\rangle$ (where $\mathbf{a}$ is the lattice constant). The cyan Burgers circuits further confirm dissociation of the full dislocation into two partial dislocations, each with Burgers vector $\frac{1}{2} \mathbf{b} = \mathbf{a}\cdot\langle 011\rangle$.

Regarding partial dislocation-domain wall interactions, we fully agree that such interactions can impede dislocation glide and thus influence plastic deformation in perovskites. For CsPbBr₃, synthesized at high temperatures in cubic phase, cooling to room temperature induces phase transformations from cubic to tetragonal to orthorhombic¹⁵. This cooling process can introduce domains with different orientations due to asymmetric [PbBr₆]⁴⁻ octahedral tilts along $[001]_{pc}$ and $[010]_{pc}$, where “pc” denoted pseudo cubic. As reported in prior study¹⁶, in bulk crystals, these domain walls appear as sharp-contrast lines in low-magnification BF-TEM images (Fig. R7a), which resembles twin boundaries, supporting by the corresponding SAED pattern that contains two zone axis in Fig. R7b. However, our experiments used single-crystalline

micropillars, as shown in Fig. R7c, the deformed 600 nm diameter pillar does not contain obvious domain walls like those in bulk samples, the corresponding SAED pattern (Fig. R7d) also supports single-crystalline nature. We speculate that phase transformation kinetics is different in sub-micron samples during cooling, potentially suppressing domain formation due to size constraints or reduced nucleation sites.

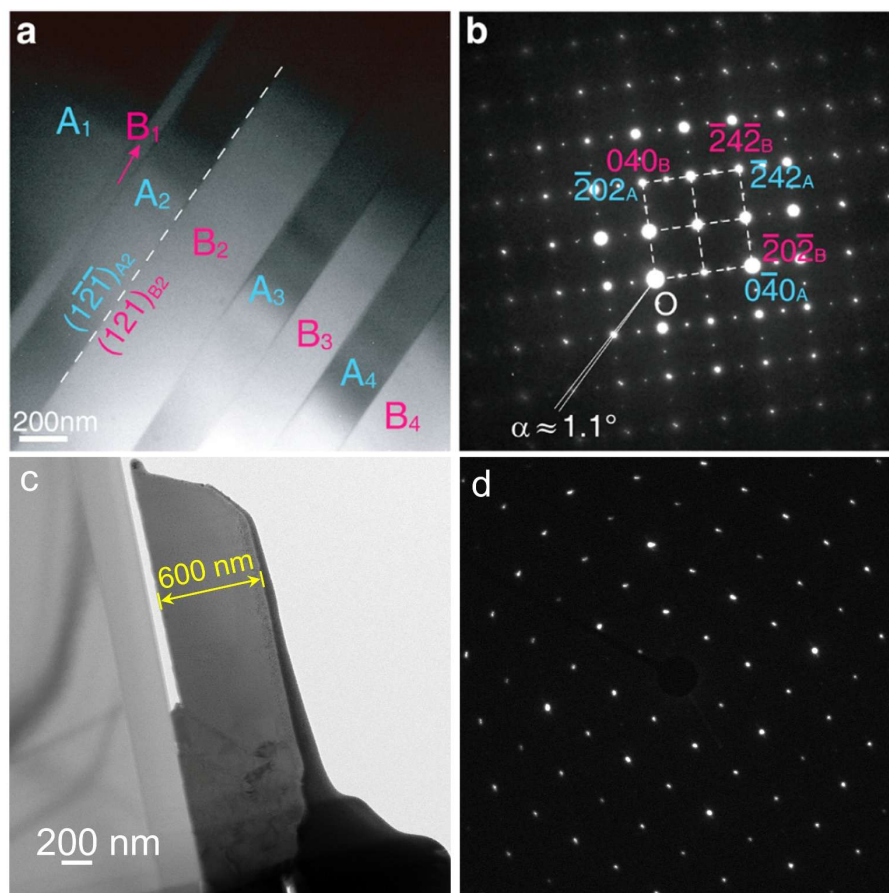


Fig. R7. Domain walls in bulk and sub-micron CsPbBr₃ samples. **a**, BF-TEM image showing domain walls in bulk CsPbBr₃ sample from ref. 16, appearing as sharp lines. **b**, Corresponding SAED pattern from domain A-B boundary in ref. 16, showing bi-crystalline. **c**, BF-TEM image of sub-micron pillar with diameter of 600 nm, showing no obvious domain boundaries like bulk sample. **d**, The corresponding SAED pattern confirms single crystallinity.

2. Given that the experiments were conducted on single crystal nanopillars, no STEM experimental characterization on configurations and reactions associated with activity related to interactions of non-coplanar dislocations

Response:

The favourable, or only possible dislocation slip system in room-temperature deformed perovskites is $\{011\}\langle 0\bar{1}1\rangle$ slip system, which has four equivalent variants under the current loading geometries (Fig. R8a). Thus, non-coplanar dislocation reactions in our

samples refer to interactions among these $\{011\}\langle 0\bar{1}1\rangle$ variants. The occurrence of such reactions largely depends on the stage of deformation.

In the early stage of deformation, the small volume of micron-scale samples allows dislocations to easily escape, creating a dislocation-starved state as proposed by Julia Greer¹⁷. The typical stress-strain curve of micropillars consists of massive stress drops that nearly down to zero caused by dislocation escape (Fig. S1). Such stage often persists until engineering strains reach ~70–80%, with little dislocation accumulation and interaction. However, continued compression shortens the sample, limiting dislocation escape and causing accumulation inside. At this stage, stress increases dramatically, and non-coplanar dislocations can react with each other, for example:

$$\frac{1}{2} \mathbf{a} \cdot [01\bar{1}] + \frac{1}{2} \mathbf{a} \cdot [0\bar{1}1] = \mathbf{a} \cdot [00\bar{1}]$$

a typical non-coplanar dislocation interaction induced dislocation wall is demonstrated in Fig. R8b.

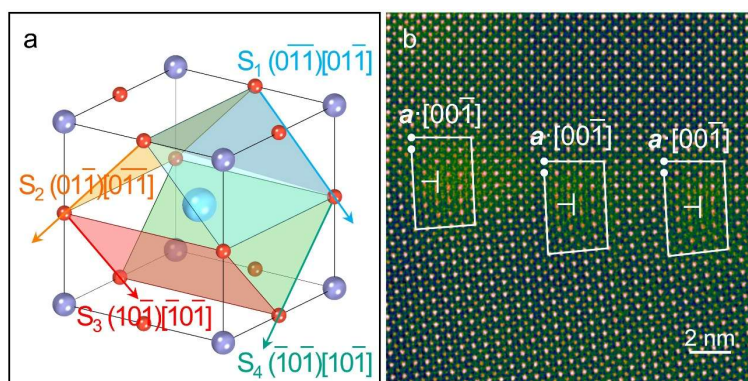


Fig. R8. Non-coplanar dislocation slip systems and their interactions. **a**, The four equivalent variants of the $\{011\}\langle 0\bar{1}1\rangle$ slip system. **b**, A $\langle 001\rangle$ dislocation wall resulting from non-coplanar dislocation interactions in the late stage of compression.

When discussing plasticity of perovskite micropillars, we typically focus on the early stage, which introduces minimal residual dislocations without introducing cracks or cleavages. Such deformation will not affect the functional properties of the materials, as detailed in our previous study¹⁴. Due to the low dislocation density in this regime, STEM characterization of non-coplanar interactions was not feasible in our samples. According to the suggestion, we've added the following brief discussion in line 136 to 141 in the revised manuscript and expanded Supplementary Note 1 to clarify potential interactions among non-coplanar dislocations:

“ $\{110\}\langle 1\bar{1}0\rangle$ systems, which are both energetically favoured in perovskite materials and geometrically favoured under the current loading orientation, as detailed in Supplementary Note 1. Most dislocations can slip out of the microscale samples, forming a dislocation-starved state⁴³, as evidenced by the serrated stress-strain curve (Fig. S1) correlating with dislocation nucleation and glide events. Such characteristics can prevent non-coplanar dislocation interactions.

3. A discussion on the nature of primary and secondary slip planes is missing and linking schmid factor analysis would make the experimental work far more compelling

Response:

We thank the reviewer for this valuable suggestion. As discussed in our response to your comment #1, the $\{011\}\langle 0\bar{1}1\rangle$ slip system is universally the primary slip system in perovskites. It is not only the most energetically favourable among common low-index slip systems, as evidenced by the generalized stacking fault energy profiles in Fig. R9 from our previous work¹⁴, but also exhibits the highest Schmid factor under the current loading geometry along the $[00\bar{1}]$ orientation. As shown in Table R4, all the four equivalent variants of the $\{011\}\langle 0\bar{1}1\rangle$ system achieve the maximum Schmid factor of 0.5.

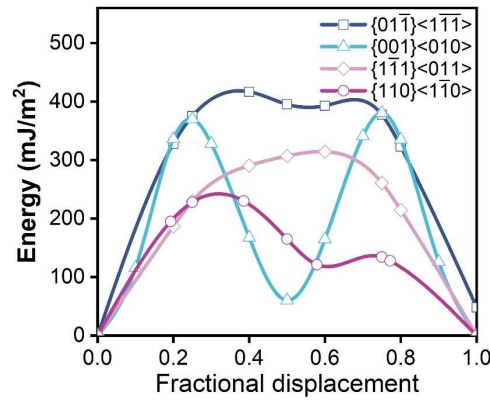


Fig. R9 Generalized stacking fault energy (GSFE) profiles of slip systems in CsPbBr₃.

In contrast, though secondary slip systems (e.g., $\{01\bar{1}\}\langle 1\bar{1}1\rangle$ or $\{1\bar{1}1\}\langle 011\rangle$ have a slightly lower Schmid factors of 0.41, they are rarely observed at room-temperature deformation, due to the large slip energy barrier induced by ionic repulsion. The related discussion on primary and secondary slip planes, including the Schmid factor analysis in Table R4, have been added and highlighted in Supplementary Note 1. This enhances the context of our experimental observations and strengthens the link between the observed dislocation structures and the underlying deformation mechanisms. We believe this addition makes the work more compelling while maintaining focus on the core findings.

Table R4 The Schmid factors of slip systems when loaded along $[00\bar{1}]$ direction.

Loading direction	Slip system	Schmid factor
$[00\bar{1}]$	$(0\bar{1}1)[01\bar{1}]$ (denoted as “S ₁ ”)	0.5
	$(01\bar{1})[0\bar{1}1]$ (denoted as “S ₂ ”)	
	$(10\bar{1})[\bar{1}01]$ (denoted as “S ₃ ”)	
	$(\bar{1}01)[10\bar{1}]$ (denoted as “S ₄ ”)	
	$\{01\bar{1}\}\langle 1\bar{1}1\rangle$	0.41
	$\{1\bar{1}1\}\langle 011\rangle$	0.41
	$\{001\}\langle 010\rangle$	0
	$\{001\}\langle 110\rangle$	0

Reviewer #3 (Remarks to the Author):

Summary: The authors examined the effect of a pulsed and continuous electron beam on glide dislocations, claiming that the pulsed beam induces glide dissociated dislocations and continuous causes glide-to-climb dislocations. They also make the claims that the local chemistry governs the climbing mechanism when using the continuous electron beam.

Overview: First of all, the authors should be proud of their ability to visualize dislocation translation so clearly and prescribing the motion to chemical and strain based analysis, which are also accompanied by great simulations. Unfortunately, the most important aspect of the paper “changing the electron beam from pulsed to continuous translates the dislocations” has numerous flaws which interfere with the rest of the manuscript. The author’s biggest claim is that the pulsed electron beam keeps the dislocation steady, but then turning off the chopping modulator displaces it. The motion is clear in the data, but the interpretation and experimental descriptions do not support the data being presented and the experimental techniques contradict their explanations of the results. Specifically looking at Figures 2 and 3, the methods shows the pulsed beam as repetitive which means that in a single image, the time-resolved data points would be compressed into a single data point when the frequency is higher (authors say “high frequency”) than the refresh rate of the detector. If the authors were not using a frequency-based approach, but instead, a triggered approach, then the results would be achievable with electrostatic delays between the EDM and detector. Additionally, the crucial data sets being compared have many problematic differences, particularly a 40x dose increase which is ignored as a possible cause for the transition. The authors only prescribe the change to the pulsing aspect, but a 40x change in dose seems very large to ignore. These problems make me believe that the experiments have not been performed with care and need to be reevaluated for accuracy. As such, I cannot suggest the manuscript for publication at this point until the technical details of the experiments are well polished. However, if the authors can create consistency between the experiments performed and data collected while also conducting more control experiments to verify whether these results are dependent on dose, pulsed/continuous, rest time, etc. there is massive interest because electron beam induced dynamics are an extremely contentious and interesting topic, not to mention that Figure 3c-e contains remarkable data!

Response:

We are truly honored to have received your enthusiastic recognition of the significance of our work. We are especially grateful for your critical and constructive comments, which have greatly helped us improve the rigor and presentation of the manuscript and substantially enhanced the overall quality of this study. Below are our point-by-point responses.

Major Comments:

The authors need to provide line numbers for easier review process, comments below will be difficult to track because they are missing.

1) Sentences relating to references 26-28 – These claims are out of date and I wouldn't use language that implies it is impossible for those techniques. The truth is that they are already achievable, but NOT easy. You should focus on why this technique is better than those below. Your images are higher resolution and any distortions don't appear present, but implying that other techniques cannot get atomic resolution is incorrect as provided in the references below:

Laser-Driven Pulsed Beam - Pavel K. Olshin, et. al., Nano Letters 2021 21 (1), 612-618, DOI: 10.1021/acs.nanolett.0c04184

RF Cavity Driven Pulsed Beam - Spencer A. Reisbick, et. al., , Ultramicroscopy, Volume 235, 2022, 113497, ISSN 0304-3991, <https://doi.org/10.1016/j.ultramic.2022.113497>.

Pulsed STEM imaging - Thomas E Gage, et. al., Microscopy and Microanalysis, Volume 30, Issue Supplement_1, July 2024, ozae044.723, <https://doi.org/10.1093/mam/ozae044.723>

Response:

We thank the reviewer for this constructive comment, which has helped us refine the discussion on pulsed-beam techniques to better reflect the current state of the field. Following the suggestion, we have revised the relevant sentences to avoid implying that atomic resolution is impossible with laser-driven and RF methods, acknowledging instead that recent advances have enabled atomic-resolution (~ 2 Å) imaging with these techniques, and incorporated the first two provided references as refs. 32 and 33 in the revised manuscript. Additionally, we have cited the third reference as ref. 35 in the revised manuscript and clarified it as electrostatic-deflection-based method alongside our EDM-based pulsed beam (newly added ref. 34), both relying on rapid blanking of a continuous beam without compromising intrinsic beam properties. These revisions have significantly strengthened the manuscript by providing a more accurate technological background. The following revision is highlighted in lines 86 to 94 of the revised manuscript:

“While traditionally limited in spatial resolution due to increased energy spread and space charge effects (e.g., Boersch effect), recent instrumental advances have enabled atomic-resolution imaging at ~ 2 Å in some implementations^{32,33}. Another emerging strategy employs electrostatic deflection to rapidly blank or chop a continuous beam into pulses^{34,35}, exemplified by electrostatic dose modulator (EDM)³⁴. This approach operates without altering source properties like spatial and temporal coherence. When integrated with aberration-corrected TEM, EDM significantly reduces electron dose rates while maintaining the pristine sub-angstrom space resolution of TEM.”

2) 4 th paragraph in introduction – There is no mention of how the EDM system works. Is it a fast chopping method? Does it sweep the beam? What signals does it use to chop? Is it chopping using a mechanical block or an electrostatic method? What is the “high frequency”? There either needs to be a reference describing how the pulsing occurs or it needs to be described in this paper (or SI). So far, there has been significant text dedicated to explaining why this EDM system is a better method for these beam sensitive materials, without describing why, and there is no reference to it provided.

Response:

Thank you for pointing out the lack of description on the EDM system’s operating principles. We agree that a detailed explanation is essential to substantiate why EDM is advantageous for beam-sensitive materials like perovskites. In response, we have added a reference to the EDM technology [Microsc. Microanal. 2022, 28, 2230] as ref. 34 in the revised manuscript. The basic working principles and introduction can also be found on the JEOL official website: <https://www.jeol.com/products/scientific/tem/EDM.php>, here we reproduced it in Fig. R10 to clearly illustrate.

[Figure Redacted]

Fig. R10. Schematic of the working mechanism of the electrostatic dose modulator (EDM), reproduced from the JEOL official website
(<https://www.jeol.com/products/scientific/tem/EDM.php>)

The EDM system employs a fast electrostatic chopping (blanking) method, distinct from mechanical block. It is placed pre-sample and rapidly deflects the continuous electron beam generated by the TEM’s electron gun. The EDM simply blanks the beam on and off without spatially sweeping it. The “high frequency” refers to the switching

rate, with a maximum pulse frequency of 500 kHz. While this does not match the temporal resolution of ultrafast TEM, it adequately enables synchronization of the pulse period with STEM pixel dwell times (e.g., 4 μ s in our experiments), facilitating pixel-by-pixel dose modulation (the details will be shortly illustrated in the response to your comment #3). EDM also supports seamless in situ switching between pulsed and continuous modes without realignment or focus changes which is critical for tracking dynamic beam-induced transitions in perovskites at atomic resolution, as demonstrated in our results. These features ensure distortion-free imaging and adaptive spatiotemporal dose control.

The revisions are highlighted in lines 92-97 of the introduction, we have also expanded the Methods section with a comprehensive overview of the EDM mechanism. We believe these additions fully address the concern and enhance the manuscript's clarity and rigor.

3) Figure 2 – The motion of the dislocation before and after the application of the continuous beam seems obvious and is very interesting, but I am having a lot of difficulty translating how they are converting the data into the time-resolved plots. 3a)

a. First, the dwell time is 4 μ s, and the acquisition time is in seconds. However, the authors are claiming they can extract different time points on their data from a single image. Based on the technique they have explained, they are summing the entire 4 μ s into one frame which is part of a 5 second image (16 for continuous). So, it does not make sense how it is possible to extract each of the data points in the two plots. The authors need to provide the evidence and description of how these can be made, because the description in their methods section says these graphs should not be possible.

Response:

Thanks for pointing out the lack of clarity in our original manuscript, we reviewed our presentation and agree the experimental details are not stated clearly, which led to misunderstanding. Please allow us to clarify the experimental timeline and data analysis procedure.

First, we used STEM mode to obtain images. The electron probe visits pixels sequentially in a standard raster scan pattern: from left to right along each horizontal line, and line-by-line from top to bottom, as indicated by the red arrows in Fig. R11. In our experiments, the dwell time of electron probe on each pixel was set to 4 μ s. Under the conventional continuous beam condition, each illuminated pixel (e.g., the red pixel in Fig. R11) is irradiated for the full 4 μ s when acquiring an frame. For the pulsed beam case, we set the pulse period to 4 μ s to synchronize it with pixel dwell time. We set the pulse beam to a 25% duty cycle, this results in 1 μ s beam-on followed by 3 μ s beam-off within each pulse period/pixel dwell time. Thus, the dose rate modulation achieved with our EDM pulsed beam is on a pixel-by-pixel basis.

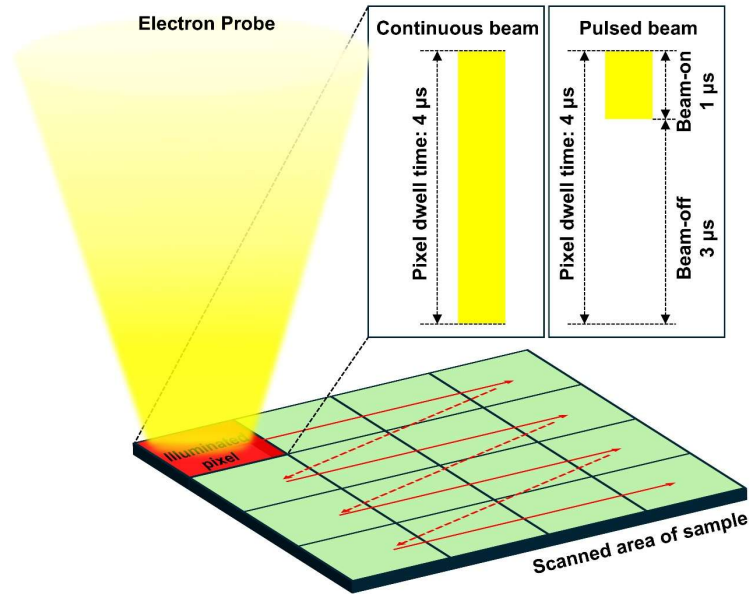


Fig. R11. Schematic illustration of pulsed and continuous beam conditions in STEM imaging. The electron probe scans the sample pixel by pixel in a standard raster pattern, with a dwell time of 4 μs per pixel. Under continuous beam, the beam-on time is 4 μs per pixel dwell time. Under pulsed beam (4 μs pulse period, 25% duty cycle), the beam is on for 1 μs followed by 3 μs off during each pixel dwell time, enabling pixel-by-pixel dose modulation.

Secondly, the full frame of images was acquired at size of 1024×1024 pixels in Figs. 2c-d and Supplementary Movie 1, and we cropped the region of interest for clearer presentation. Therefore, the acquisition time for a single frame is $1024 \times 1024 \times 4 \mu\text{s} = 4194304 \mu\text{s} \approx 4.19 \text{ s}$ per frame.

Thirdly, the plots of point defect concentration-time relationships shown in Figs. 2f and 2g as well as Figs. 3 h and 3k reflect the defect concentration evolution of a single illuminated pixel within the 4 μs pixel dwell time. With the detailed point defect generation and recombination analysis can be seen in Supplementary Note 5. This approach is valid because, during the acquisition of a whole frame of image, the electron probe visits each pixel only once, and the actual electron-sample interaction occurs exclusively within that pixel dwell time.

According to the suggestion, we now added more detailed explanation in the revised manuscript for better illustrating the imaging conditions and setup. The following revisions were made in the revise manuscript:

Line 161-166: “To achieve pixel-by-pixel dose rate control during STEM imaging, the pulsed beam period was synchronized with the electron probe’s dwell time on each pixel, both set to 4 μs. Under the setup of 25% duty cycle, the beam is on for 1 μs and off for 3 μs within each 4 μs pixel dwell time/pulse period. Consequently, the dose rate of pulsed beam is reduced to $\sim 2.86 \cdot 10^{10} \text{ e}^- \text{ nm}^{-2} \text{ s}^{-1}$, which is 25% of that of the original continuous beam”

Line 175-178: “Furthermore, close the EDM enables seamless in situ switching of pulsed electron beam to a continuous beam, of which the dose rate increases fourfold

to $\sim 1.14 \cdot 10^{11} \text{ e}^- \text{ nm}^{-2} \text{ s}^{-1}$ compared with pulsed condition, without the need for realignment or focus adjustment”

Line 208-211: “Here, we evaluate the defect concentration dynamics during the electron probe’s dwell time on a single pixel. This approach is valid, as the probe visits each pixel only once during the acquisition of an entire image frame, with electron-sample interactions occurring exclusively within each pixel’s dwell time.”

b. Second, the authors are claiming that the pulsed vs continuous triggers the transition, however the only parameter which stays consistent between the two experiments is dwell time. I would assume that the 40x (1.1×10^{10} to 4.57×10^{11}) dose increase is likely to have a major impact as well. I was very disappointed to find that there were no careful controls taken to verify that the pulsing nature is the sole cause for the transition.

Response:

We agree with the reviewer and apologize for not carefully controlling the variables, which has also been indicated by Reviewer #1 in comment #2. As also detailed in our response to that comment, we pitifully employed larger magnification and image size of 2048*2048 for acquiring images under continuous beam in the dataset of original Fig. 2e, which induces extra larger electron dose rate.

Therefore, we conducted new experiments with all the parameters are well-controlled. All the imaging parameters for pulsed beam and continuous beam are same, except the on and off status of EDM, the parameters are listed in Table R5. Under such setup, the dose rate of continuous beam is exactly 4x of pulsed beam with 25% duty cycle. The newly collected results are presented in Fig. R1 and the updated Fig. 2 in revised manuscript.

Besides, we conducted new experiments that deconvolved the effects of dose rate, total dose and pulsing nature, the detailed discussion can be found in the response to the 2nd comment of Reviewer #1, Table R1, Figs. R1-4. The results rule out total accumulated dose as the primary driver for climb, and unambiguously demonstrated that the pulsing nature, which not only minimizes dose rate, but also provide beam-off intervals for periodic recovery, is the key to maintaining intrinsic dislocation structures. A discussion decoupling these effects has been added into revised manuscript, as can be seen in line 246-257.

Table R5 Beam parameters for pulsed and continuous beam conditions in strictly controlled HAADF-STEM experiments.

Beam	Pixel dwell time (μs)	Pulse period (μs)	Beam-on time on each pixel (μs)	Dose rate ($\text{e}^- \cdot \text{nm}^{-2} \cdot \text{s}^{-1}$)	Dislocation structure
Pulsed	4	4	1	$2.86 \cdot 10^{10}$	glide
Continuous	4	NA	4	$1.14 \cdot 10^{11}$	climb

c. Third, when performing STEM imaging, a small portion of the sample receives a lot of dose, then the beam moves to the next location. For the parameters provided (4 μ s dwell time, 4.5 s acquisition), this means that the region of interest is illuminated by the electron for 1 μ s (4 μ s for continuous), then nonilluminated for $4.5\text{ s} - 1\text{ }\mu\text{s} = 4.49999\text{ s}$ (4.49995 s for continuous). Even if we assume the region of interest is 10% of the size of the image (huge overestimate), the interaction time between the defect and the electron beam is still less than 0.001% per frame. In this figure, they are implying that there is no down time for the defect, but that is not how STEM works. Does this mean that not being on the dislocation causes it to move, and if so how are you able to tell until the beam arrives at that location?

Response:

Yes, in STEM mode, point defect generated within an illuminated pixel largely recombine and annihilate once the probe moves to other pixels. As stated above, we plotted and compared the point defect concentration evolution within a single 4 μ s pixel dwell time under both pulsed beam and continuous beam.

In the newly added dataset (updated Fig. 2, also presented in Fig. R12), the peak defect concentration C_0 introduced by pulsed beam (Fig. R12a) reaches only about half the value induced by continuous beam (Fig. R12b). Moreover, under pulsed conditions, C_0 is sustained for only a very short duration of $\Delta t_s = 0.54\text{ }\mu\text{s}$. Immediately afterward, during the beam-off period starting at $t = 1\text{ }\mu\text{s}$, point defects undergo rapid recombination and annihilation. In contrast, the continuous beam introduces twice the peak defect concentrations than pulsed beam case, and maintains this elevated level for a much longer sustained duration of $\Delta t_s = 3.77\text{ }\mu\text{s}$. The combination of higher defect concentration and significantly longer sustained duration provides sufficient driving force to activate dislocation climb within the pixel dwell time.

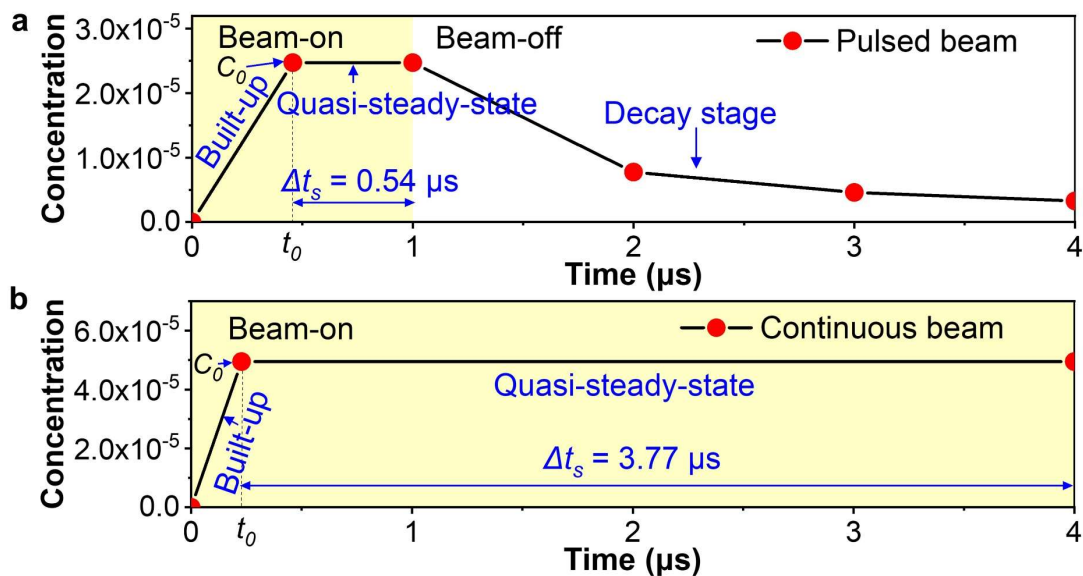


Fig. R12. Point defect concentration dynamics during a single 4 μ s pixel dwell time under pulsed and continuous beam conditions. a, Pulsed beam condition described in Fig. 2c. The curve consists of an initial build-up stage, followed by a quasi-steady-state with peak concentration $C_0 = 2.47 \cdot 10^{-5}$ reached at $t_0 = 0.46\text{ }\mu\text{s}$ and sustained $\Delta t_s = 0.54\text{ }\mu\text{s}$ until beam-off

at $t=1\ \mu\text{s}$, after which recombination dominates and concentration decays. **b**, Continuous beam described in Fig. 2e. The curve shows rapid build-up to a higher peak concentration $C_0 = 4.95 \cdot 10^{-5}$ at $t_0 = 0.23\ \mu\text{s}$, followed by a quasi-steady-state that persists for the remainder of the dwell time ($\Delta t_s = 3.77\ \mu\text{s}$).

4) Figure 3 – When examining Figure 2, the data quality between the continuous and pulsed electron beam, I can kind of see where the similarities in the sample are. However, in Figure 3, the panels for (f) and (i) do not even look like they are in the same area. The authors should explain why the sample area looks so different. Additionally, the difference between this and Figure 2 is the dose of the pulsed beam, so the authors should elaborate more on why the dose causes the difference in the translation.

Response:

The images in Figs. 3f and 3i were acquired from the same location, aligned using characteristic features in a larger field of view (Fig. R13). The apparent difference between them is primarily due to the very low signal-to-noise ratio in Fig. 3f (Fig. R13a).

Imaging the perfect intrinsic glide-dissociated dislocation in SrTiO_3 is extremely challenging because of its high beam sensitivity (especially for oxygen). As detailed in our response to Reviewer #1 (Comment #4) and Supplementary Note 5, we balanced the need for sufficient signal (higher dose rate) against the need to preserve the intrinsic structure (lower dose rate). Even at the minimum dose rate required to resolve the lattice, climb was initiated. Consequently, the best observable state was the “near-glide” configuration in Fig. 3f, which is a transitional state between ideal glide and climb dissociation. We have clearly stated this in the revised manuscript in line 285-288 and line 296-300.

Finally, the difference in image quality between CsPbBr_3 and SrTiO_3 under similar electron dose conditions (e.g., Fig. 3f and Fig. S4a in updated Supplementary Materials) arises from their distinct lattice constants ($\sim 0.583\ \text{nm}$ for CsPbBr_3 vs. $\sim 0.390\ \text{nm}$ for SrTiO_3). The larger interatomic spacing in CsPbBr_3 enables clearer resolution of its atomic columns.

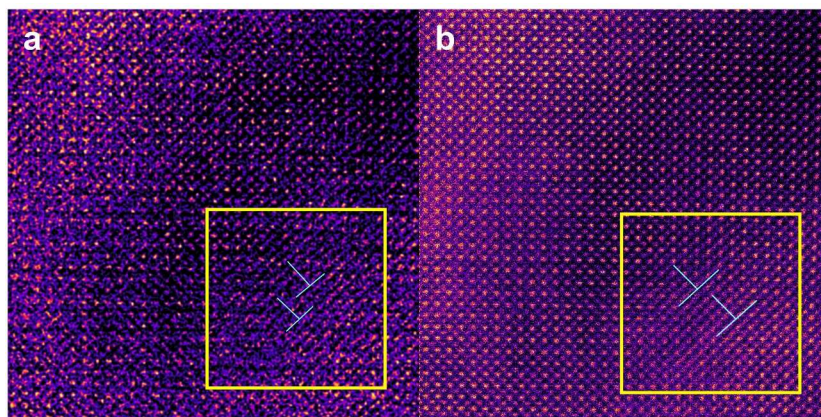


Fig. R13. Larger field of view of the dislocation region in SrTiO_3 used for alignment and comparison. a, The image corresponding to Fig. 3f (pulsed beam, near-glide configuration). **b**, The image corresponding to Fig. 3i (continuous beam, climbed configuration).

5) General Reproducibility – In searching through the supplementary information, it appears that there are no additional statistics or verification that these results can be duplicated and controlled. Especially when comparing Figure 2 and 3, the plot is the only thing that looks repeatable, but as mentioned in (3) the authors never describe how these plots work.

Response:

Thanks for the suggestion on reproducibility, with the newly added experiments in updated Fig. 2 and Fig. S5, we believe the reproducibility issue is solved. As discussed in the response to your comment #3)-a, the plots are the defect concentrations evolutions within a single pixel dwell time. As details can be found in Supplementary Note 5, the plots mainly consist of: (1) Build-up stage, when point defect generation dominates and the concentration is proportional to time with a coefficient K_0 ; (2) Quasi-steady-state, when the vacancy-interstitial recombination balanced out generation, reaching a steady and peak concentration of C_0 ; (3) Decay stage, when beam-off, no new defects are generated, only vacancy-interstitial recombination happened.

Besides, though the original Fig. 2 failed in strictly controls the variables when transferring from pulsed to continuous beam, the stable imaging of glide-dissociated dislocation structure under pulsed beam is indeed significant in uncovering the intrinsic dislocation structure, and its rapid glide-to-climb transition under continuous beam with extremely high dose rate unambiguously indicated the beam induced dislocation climb behaviour. Therefore, we relocated the dataset into to Supplementary Note 6 as Fig. S4 and clearly stated the imaging conditions.

Minor Comments:

1) The authors need to provide line numbers for easier review process, comments below will be difficult to track because they are missing.

Response:

We thank the reviewer for this helpful suggestion. To facilitate the review process and make it easier to track our changes, we have added continuous line numbers throughout the revised manuscript.

2) Introduction – Paragraph 1 – Sentence 2 – “That” seems like it isn’t supposed to be here.

Response:

We appreciate the reviewer identifying this grammatical error. We have corrected the sentence by replacing the erroneous “that” with “, which are” for proper relative clause structure. The change is highlighted at line 58 in the revised manuscript.

3) Resolving dislocation structures in CsPbBr₃ by EDM-based pulsed electron beam – Paragraph 1: This paragraph seems nearly word-for-word a repeat of the introduction. I even thought that I accidentally went backwards in the manuscript it was so similar.

Response:

Thanks for highlighting the unnecessary repetition with the Introduction. To address this repetition, we have significantly shortened the opening paragraph in the section “Resolving dislocation structures in CsPbBr₃ by EDM-based pulsed electron beam” and merged it with the subsequent paragraph for better flow (see lines 155-161 in the revised manuscript). These changes eliminate redundancy while retaining the essential justification for our experimental approach. The revised text now reads:

“Conventional continuous electron beams (Fig. 2a) deliver high dose rates during atomic-resolution STEM imaging, raising concerns regarding potential beam-induced structural modification¹⁵. To address this, we employed a programmable pulsed electron beam generated by an EDM (Fig. 2b), which electrostatically deflects the continuous beam into pulses without altering beam properties, preserving native sub-angstrom resolution while substantially reducing dose rates and mitigating irradiation effects.”

4) The figures are very difficult to follow. There needs to be more description in the captions and labels on the panels. One example is Figure 3, but this is true for all figures.

Response:

We thank the reviewer for this important suggestion. To improve clarity, we have substantially expanded all figure captions to provide detailed descriptions of each panel, including experimental conditions, key observations. Additionally, we have enhanced in-figure labels and added clear panel designations. Corresponding descriptions in the main text have also been revised and expanded to better guide readers to the key features in each figure, as highlighted in the related contents. These changes made the figures much easier to follow independently.

With these new experiments and explanations, we believe the concerns are well-addressed. We’d like to take this opportunity to thank you for all the constructive comments, which truly helped us a lot to present our work in a more rigorous and clear way. Thank you.

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Responses to Reviewers' Comments (NCOMMS-25-85515A)

Below are our point-by-point responses (shown in **black** text) to the reviewers' comments (shown in **blue** text).

Reviewer #3 (Remarks to the Author):

Overall, the manuscript is in a significantly improved version, with the only remaining concern being the ways the time resolved panels (Figures 2g, 2h, 3h and 3k) are construed. I understand that each frame is collected in a 4D STEM configuration where each pixel receives 4 μ s (1 μ s) of dose before experiencing $\sim$ 4s of "darkness" in the continuous (pulsed) configuration. I also understand that these plots requires three parameters, K_0 , K_{iv} and t , such that K_{iv} and t are a constant and the variable, respectively. This leaves only K_0 , which dC/dt or the change in concentration with time. The only data which changes in time is the frame-by-frame acquisition of the images (because the 4 and 1 μ s dwell times are all compressed into 1 frame of $\sim$ 4s). This means that K_0 is a measure of different frames. Therefore, the panels in question are calculations solely made from the measurement between multiple frames. However, they are presented as if each data point is collected independently, not calculated from the C_0 point. I have no problem with the calculations which are made or the results they are describing, but find it deceptive to illustrate these graphs as if they were measurements. However, if there are details about this that I am missing, then this section needs significant clarity on the data collection and figure creation.

Response:

We sincerely thank the reviewer for the recognition of our revised manuscript and the constructive comments which have facilitated the enhancement. We greatly appreciate the continued careful reading and expertise invested in evaluating our manuscript.

Thanks for highlighting the need for clearer clarification regarding the data presentation in the time-resolved point defect concentration panels. Indeed, the curves in question represent analytical solutions derived from standard radiation damage rate equations (as described by Gary S. Was, *Fundamentals of Radiation Materials Science*, Springer, 2007), rather than direct experimental measurements. In the previous submission, we presented the defect concentration dynamics with discrete key data points (Fig. R1a), which likely contributed to the misleading impression that these were measured. To eliminate any potential ambiguity, we have now replaced the discrete data points with continuous analytical curves generated directly from the solution of the rate equations (Fig. R1b; the corresponding values have also been updated, as detailed in the response to the next comment). All relevant figures depicting time-resolved concentration dynamics have been updated accordingly to prevent misinterpretation.

In addition to the graphical revisions, we have incorporated explicit clarifications in the revised manuscript:

Main text (lines 209-211): “Based on the standard rate equation for irradiation-induced point defect⁵⁰, the time-resolved defect concentration dynamics can be analytically calculated”.

Supplementary materials (Note 5): “This note reveals the calculated analytical solution of dynamics of point defect concentrations in CsPbBr₃ and SrTiO₃ perovskites under electron beam irradiation based on the rate equations¹²”.

Figure captions: All the related captions were revised to state that the panels depict the calculated evolution of defect concentration.

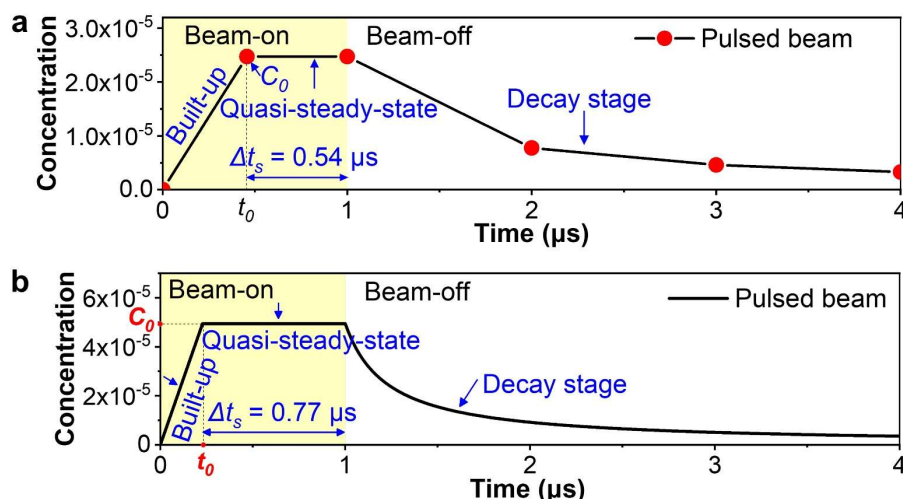


Fig. R1 Time-resolved defect concentration dynamics. **a**, Curve presented in the previous manuscript, constructed using discrete data points. **b**, Revised curve, plotted as the continuous analytical solution of the rate equations.

The only issue with the scientific content is the long “darkness period” and the assumption that there are no changes within the sample during this time. Even though the pulsed versus continuous beam has a 25% duty cycle, the actual duty cycles on a per-pixel basis is $1\mu s/4.19s = 2.4 \times 10^{-7}$ and $4\mu s/4.19s = 9.5 \times 10^{-7}$. I am surprised to see the differences in the “on” times, specifically when comparing all the time-resolved figures. In Figure S6 and 3, the build up is similar, which I would expect. However, when I look at Figure S4 and the others (Figure 2), the build up times can be extremely different between the pulsed and continuous. These differences suggest that the build up phase may not be consistent between experiments which causes the assumption to be worrisome. Therefore, I believe an explanation discussing the build up discrepancies or possibilities of “dark” dynamics during the beam off periods as one of these would give explanation to this concern.

Response to “dark” dynamics during the beam off periods

Thanks for invoking the discussion of defect dynamics during the long inter-frame “dark” periods. We have carefully considered whether undetected structural changes could occur during these intervals.

Taking Fig. 2 as an example, the point defect concentration and the sustained period of peak concentration ($\Delta t_s = 3.77 \mu\text{s}$) induced by the continuous beam are sufficient to trigger dislocation climb. As shown in Figs. 2e-f and Supplementary Movie 1 (frames 27-31), climb proceeds incrementally frame by frame. Critically, once climb has initiated (even just by a single lattice unit), the resulting configuration adopts a lower-energy state relative to the pristine glide-dissociated structure. This is supported both by our own MD simulations (Fig. R2; see also Fig. S13d) and by previous reports (*Acta Mater.* 2025, 285, 120636), rendering the climb process irreversible. During the subsequent long “dark” period, any residual free point defects undergo rapid recombination, effectively resetting the mobile defect population before the next pixel scan; the post-climb configuration, however, remains unaffected until the next illumination cycle therefore advances the climb process further, consistent with the progressive climb observed in the movie.

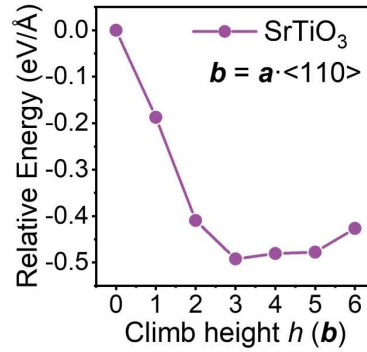


Fig. R2 Energy profile of a dislocation as a function of climb height.

For the pulsed beam condition, the sustained period of peak concentration is substantially shorter. This brief on-time is insufficient to initiate a detectable climb event. Once the beam is gated off, the generated defects undergo rapid recombination during the beam-off period and the subsequent long inter-frame dark interval. Consequently, the glide-dissociated configuration can be maintained over the 26 consecutive frames.

We have incorporated a detailed explanation of these dark-period dynamics into the revised manuscript accordingly (see lines 182-185, and lines 247-254). We thank the reviewer for prompting this clarification, which strengthens the mechanistic interpretation of our observations.

Response to different build-up stages between pulsed and continuous beams

The reviewer’s acute observation regarding the apparent discrepancies in the build-up stages between the pulsed and continuous beam conditions was extremely helpful. This comment inspired us to reconsider our treatment of dose rate in the manuscript.

Upon thorough re-evaluation, we identified that the electron dose rate of the pulsed beam must be carefully distinguished between the time-averaged dose rate Φ_{ave} (which accounts for the beam-off intervals), and the beam-on dose rate Φ_{on} (which considers

solely the beam-on period). For the well-controlled imaging conditions in Fig. 2, the sole difference between the pulsed and continuous beam modes is the on/off of the EDM, which simply chops/deflects a continuous beam into pulses without changing the beam (Fig. R3). Thus, Φ_{on} is identical to the dose rate of the continuous beam $\Phi_{continuous}$. The time-averaged dose rate of the pulsed beam, Φ_{ave} , however, is lowered to 25% of $\Phi_{continuous}$ due to the introduction of 75% beam-off intervals.

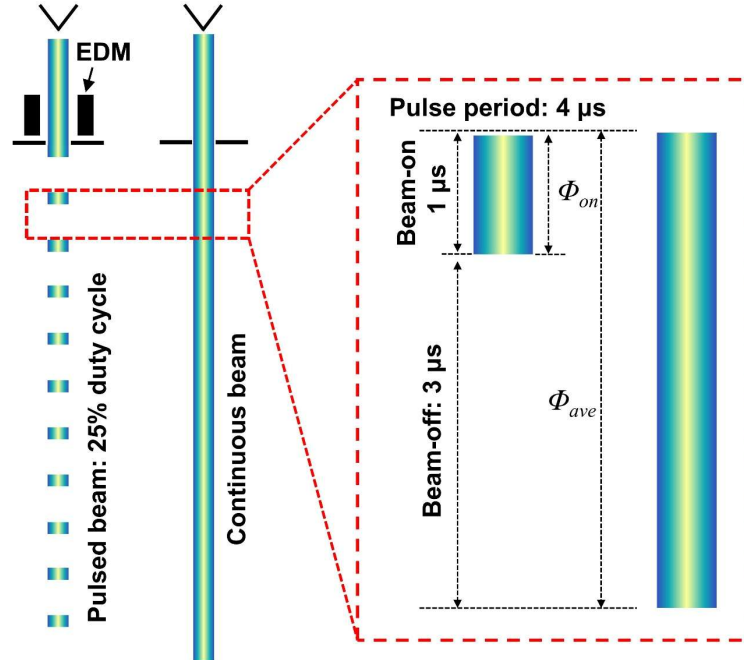


Fig. R3. Schematic illustration of the pulsed and continuous beam conditions in the well-controlled experiments of Fig. 2. The only experimental variable is the on/off state of the EDM; all other beam parameters remain unchanged.

Therefore, due to the identical Φ_{on} and $\Phi_{continuous}$, the defect generation rate K_0 , and thus the initial linear build-up stage of defect concentration should be identical for both the pulsed and continuous beams during the beam-on period (0-1 μ s). In the previous manuscript, we failed to distinguish the time-averaged dose rate Φ_{ave} from beam-on dose rate Φ_{on} and incorrectly treated Φ_{on} as 25% of $\Phi_{continuous}$, which erroneously led to distinct build-up kinetics. We have now clarified these definitions (lines 162-165), corrected the related values (line 217, 227-228, 232, 241-243), and updated the concentration-time curves in Figs. 2g-h, as presented in Fig. R4. All relevant instances of misinterpreted Φ_{on} and the associated build-up stages, together with the defect concentration dynamics curves in Fig. 3h, Fig. S4d, Fig. S6b, and Tables S4-6, have been thoroughly revised. In addition, as noted in our previous response, the datasets in Fig. 3 and Fig. S4 were acquired under uncontrolled variables (magnification and image size); consequently, the build-up stages for the pulsed and continuous beam conditions differ in those figures, which is independent to the corrections applied here.

After this further revision upon your advice, it is clearer that the primary factor preserving the intrinsic dislocation structure under pulsed beam illumination is the beam-off intervals it induces. These intervals truncate the sustained duration of peak point defect concentration and enable defect recombination before climb can be

initiated. These intervals also reduce the time-averaged dose rate and the cumulative defect population on each pixel. We have accordingly updated the abstract and related discussions (see lines 41-42 and 260-264).

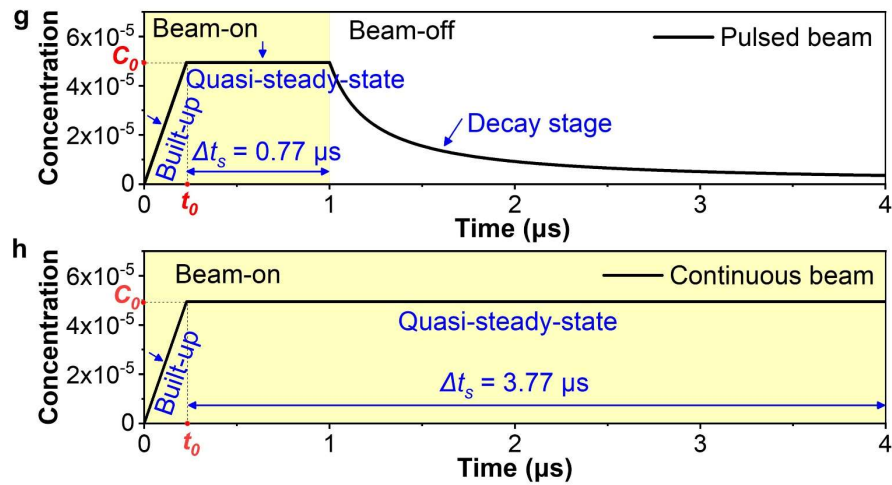


Fig. R4. Updated defect concentration dynamics under the pulsed and continuous beam conditions in the well-controlled experiments of Fig. 2. The initial build-up stages of the two curves are identical, reflecting the equal beam-on dose rate ($\Phi_{\text{on}} = \Phi_{\text{continuous}}$).

We are grateful to the reviewer for prompting us to clarify this crucial aspect, which has substantially strengthened the scientific accuracy of our work. It has been a pleasure to see the manuscript improve through this rigorous and collaborative peer-review process. We thank the reviewer once again for the thorough and insightful evaluation.

Review of Resolving intrinsic dislocation structure in perovskite crystals using pulsed electron beam with atomic resolution

Summary: The authors examined the effect of a pulsed and continuous electron beam on glide dislocations, claiming that the pulsed beam induces glide dissociated dislocations and continuous causes glide-to-climb dislocations. They also make the claims that the local chemistry governs the climbing mechanism when using the continuous electron beam.

Overview: First of all, the authors should be proud of their ability to visualize dislocation translation so clearly and prescribing the motion to chemical and strain based analysis, which are also accompanied by great simulations. Unfortunately, the most important aspect of the paper “changing the electron beam from pulsed to continuous translates the dislocations” has numerous flaws which interfere with the rest of the manuscript. The author’s biggest claim is that the pulsed electron beam keeps the dislocation steady, but then turning off the chopping modulator displaces it. The motion is clear in the data, but the interpretation and experimental descriptions do not support the data being presented and the experimental techniques contradict their explanations of the results. Specifically looking at Figures 2 and 3, the methods shows the pulsed beam as repetitive which means that in a single image, the time-resolved data points would be compressed into a single data point when the frequency is higher (authors say “high frequency”) than the refresh rate of the detector. If the authors were not using a frequency-based approach, but instead, a triggered approach, then the results would be achievable with electrostatic delays between the EDM and detector. Additionally, the crucial data sets being compared have many problematic differences, particularly a 40x dose increase which is ignored as a possible cause for the transition. The authors only prescribe the change to the pulsing aspect, but a 40x change in dose seems very large to ignore. These problems make me believe that the experiments have not been performed with care and need to be reevaluated for accuracy. As such, I cannot suggest the manuscript for publication at this point until the technical details of the experiments are well polished. However, if the authors can create consistency between the experiments performed and data collected while also conducting more control experiments to verify whether these results are dependent on dose, pulsed/continuous, rest time, etc. there is massive interest because electron beam induced dynamics are an extremely contentious and interesting topic, not to mention that Figure 3c-e contains remarkable data!

Major Comments:

The authors need to provide line numbers for easier review process, comments below will be difficult to track because they are missing.

- 1) Sentences relating to references 26-28 – These claims are out of date and I wouldn’t use language that implies it is impossible for those techniques. The truth is that they are

already achievable, but NOT easy. You should focus on why this technique is better than those below. Your images are higher resolution and any distortions don't appear present, but implying that other techniques cannot get atomic resolution is incorrect as provided in the references below:

Laser-Driven Pulsed Beam - Pavel K. Olshin, et. al., *Nano Letters* **2021** 21 (1), 612-618, DOI: 10.1021/acs.nanolett.0c04184

RF Cavity Driven Pulsed Beam - Spencer A. Reisbick, et. al., , *Ultramicroscopy*, Volume 235, 2022, 113497, ISSN 0304-3991, <https://doi.org/10.1016/j.ultramic.2022.113497>.

Pulsed STEM imaging - Thomas E Gage, et. al., *Microscopy and Microanalysis*, Volume 30, Issue Supplement_1, July 2024, ozae044.723, <https://doi.org/10.1093/mam/ozae044.723>

- 2) 4th paragraph in introduction – There is no mention of how the EDM system works. Is it a fast chopping method? Does it sweep the beam? What signals does it use to chop? Is it chopping using a mechanical block or an electrostatic method? What is the “high frequency”? There either needs to be a reference describing how the pulsing occurs or it needs to be described in this paper (or SI). So far, there has been significant text dedicated to explaining why this EDM system is a better method for these beam sensitive materials, without describing why, and there is no reference to it provided.
- 3) Figure 2 – The motion of the dislocation before and after the application of the continuous beam seems obvious and is very interesting, but I am having a lot of difficulty translating how they are converting the data into the time-resolved plots. 3a)
 - a. First, the dwell time is 4 us, and the acquisition time is in seconds. However, the authors are claiming they can extract different time points on their data from a single image. Based on the technique they have explained, they are summing the entire 4 us into one frame which is part of a 5 second image (16 for continuous). So, it does not make sense how it is possible to extract each of the data points in the two plots. The authors need to provide the evidence and description of how these can be made, because the description in their methods section says these graphs should not be possible.
 - b. Second, the authors are claiming that the pulsed vs continuous triggers the transition, however the only parameter which stays consistent between the two experiments is dwell time. I would assume that the 40x (1.1×10^{10} to 4.57×10^{11}) dose increase is likely to have a major impact as well. I was very disappointed to find that there were no careful controls taken to verify that the pulsing nature is the sole cause for the transition.
 - c. Third, when performing STEM imaging, a small portion of the sample receives a lot of dose, then the beam moves to the next location. For the parameters provided (4 us dwell time, 4.5 s acquisition), this means that the region of interest is illuminated by the electron for 1 us (4 us for continuous), then non-illuminated for $4.5\text{s} - 1\text{us} = 4.49999\text{ s}$ (4.49995 s for continuous). Even if we assume the region of interest is 10% of the size of the image (huge overestimate), the interaction time between the defect and the electron beam is still less than 0.001% per frame. In this figure, they are implying that there is no down time for

the defect, but that is not how STEM works. Does this mean that not being on the dislocation causes it to move, and if so how are you able to tell until the beam arrives at that location?

- 4) Figure 3 – When examining Figure 2, the data quality between the continuous and pulsed electron beam, I can kind of see where the similarities in the sample are. However, in Figure 3, the panels for (f) and (i) do not even look like they are in the same area. The authors should explain why the sample area looks so different. Additionally, the difference between this and Figure 2 is the dose of the pulsed beam, so the authors should elaborate more on why the dose causes the difference in the translation.
- 5) General Reproducibility – In searching through the supplementary information, it appears that there are no additional statistics or verification that these results can be duplicated and controlled. Especially when comparing Figure 2 and 3, the plot is the only thing that looks repeatable, but as mentioned in (3) the authors never describe how these plots work.

Minor Comments:

The authors need to provide line numbers for easier review process, comments below will be difficult to track because they are missing.

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Review of Resolving intrinsic dislocation structure in perovskite crystals using 1 pulsed electron beam with atomic resolution

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