

## Supporting Information

### Crystal Orientation Dependent Optoelectronic Properties in MAPbCl<sub>3</sub> Single Crystal

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## 1. Synthesis of $\text{CH}_3\text{NH}_3\text{Cl}$

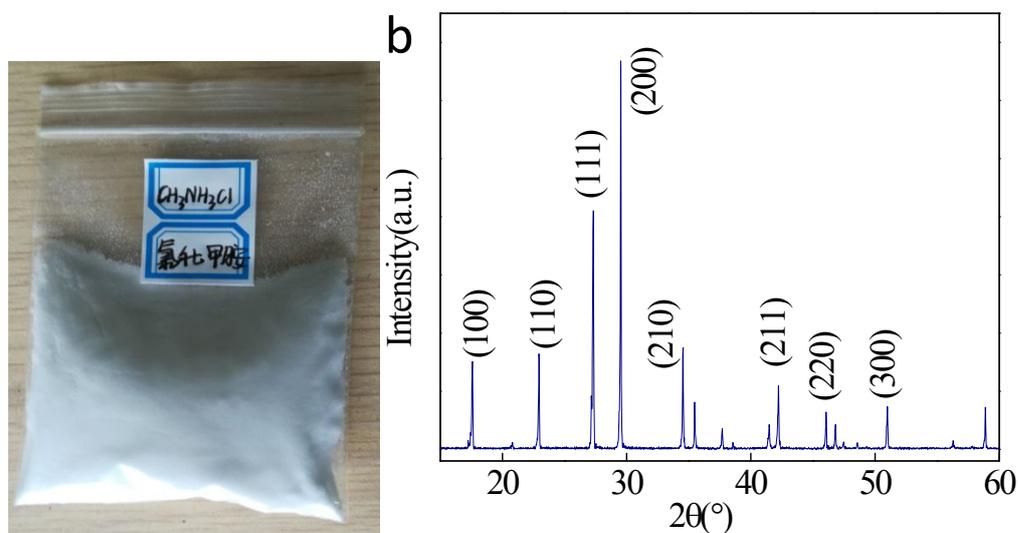


Fig. S1 (a-b) Photo and XRD pattern of synthesized  $\text{CH}_3\text{NH}_3\text{Cl}$  powder

A slight excess methylamine ( $\text{CH}_3\text{NH}_2$ ) solution was added into hydrochloric acid (HCl) solution, which was hold in an ice bath at ambient atmosphere. After reacting for more than 6 hours, the mixed solution were tightly sealed and heated at  $60^\circ\text{C}$  for 24 hours. After all the water evaporated, the white  $\text{CH}_3\text{NH}_3\text{Cl}$  crystallized. The corresponding photograph and XRD of synthesized  $\text{CH}_3\text{NH}_3\text{Cl}$  are shown in Figure S1.

## 2. Detailed time-dependent photocurrents

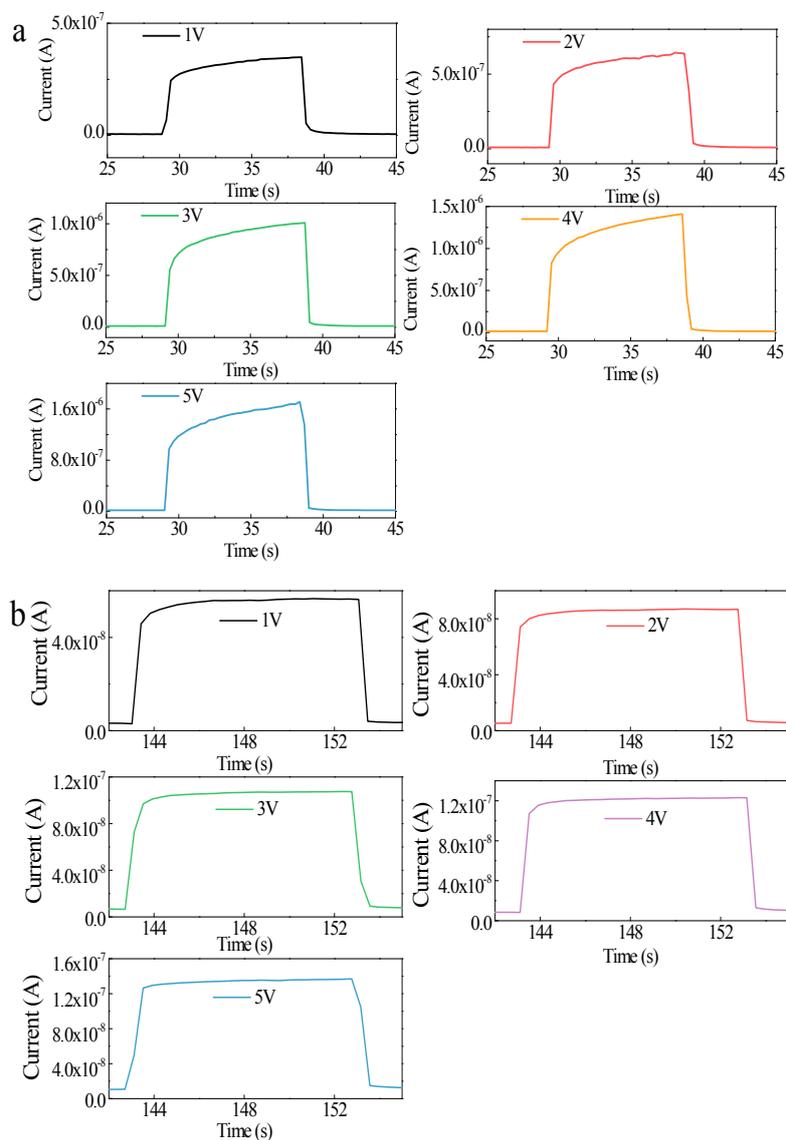


Figure S2 (a-b) Detailed time-dependent photocurrents of (100) and (110) plane devices under different voltages.

It is clear that the variation tendencies of photocurrent based on different planes is totally different. For (110) plane device, the photocurrents increase as increase of time, and they maintained stable for (110) plane device. This implies the carrier migrations for these two device is different and suggests the anisotropy play crucial roles in optoelectronic properties in MAPbCl<sub>3</sub> single crystal.

### 3. Detailed alternating $[\text{PbCl}_6]^-$ and $\text{MA}^+$ arrays

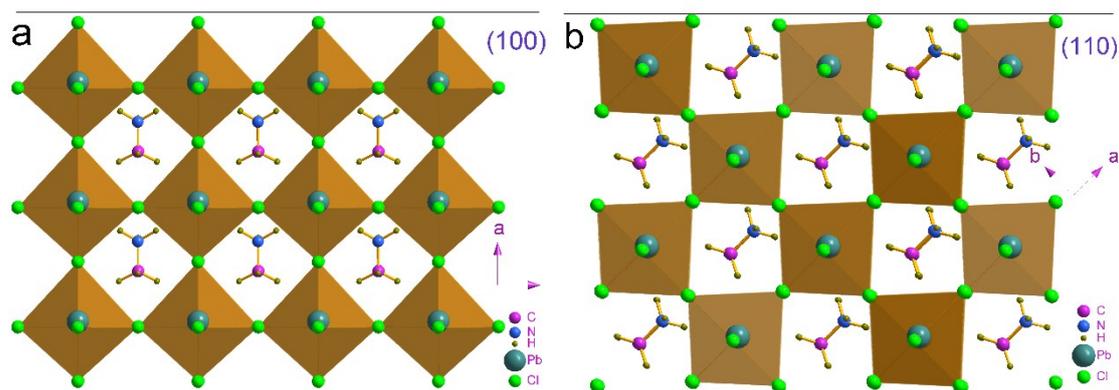


Figure S3 (a-b) Detailed alternating  $[\text{PbCl}_6]^-$  and  $\text{MA}^+$  arrays in both (100) and (110) planes

The detailed alternating  $[\text{PbCl}_6]^-$  and  $\text{MA}^+$  arrays in both (100) and (110) planes is displayed in Figure S3(a-b), from which we can see that the  $[\text{PbCl}_6]^-$  and  $\text{MA}^+$  arrangements in two planes is different. Higher distribution density of  $\text{MA}^+$  and  $[\text{PbCl}_6]^-$  in (100) plane is much easier to generate electron-hole carriers. On the other hand, higher distribution density of  $\text{MA}^+$  and  $[\text{PbCl}_6]^-$  in (100) plane also provide more  $\text{MA}^+$  and  $\text{Cl}^-$  ions carriers because hybrid perovskite  $\text{MAPbCl}_3$  is a mixed conductor. And thus the photocurrents of (100) plane is slightly higher than that of (110) plane.