Supplementary Information

Organic-inorganic Interactions of Single Crystalline Organolead Halide PerovskitesStudied by Raman Spectroscopy

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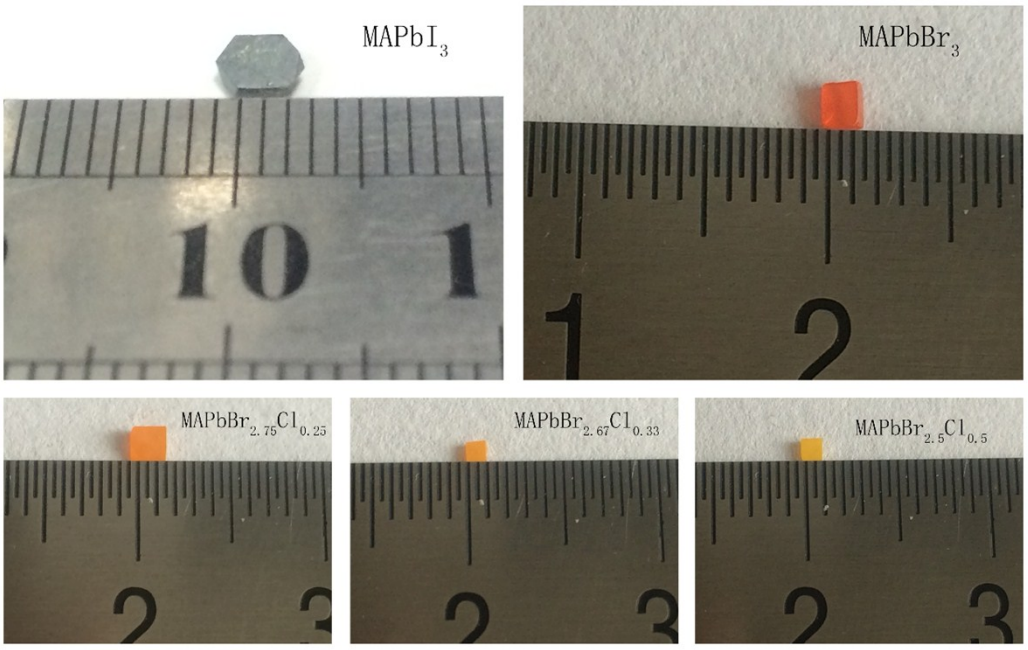

Figure S1. Photographs of various perovskite single crystals. The colors of $MAPbI_3$ (top left), $MAPbBr_3$ (top right), $MAPbBr_{2.75}Cl_{0.25}$ (down left), $MAPbBr_{2.67}Cl_{0.33}$ (down middle) and $MAPbBr_{2.75}Cl_{0.25}$ (down right) go from dark black to orange, light orange and finally become yellow. The shape of the single crystals agree with that are reported in literature[1-4], indicating that their high quality which is sufficient to our Raman experiments here although their size not so large.

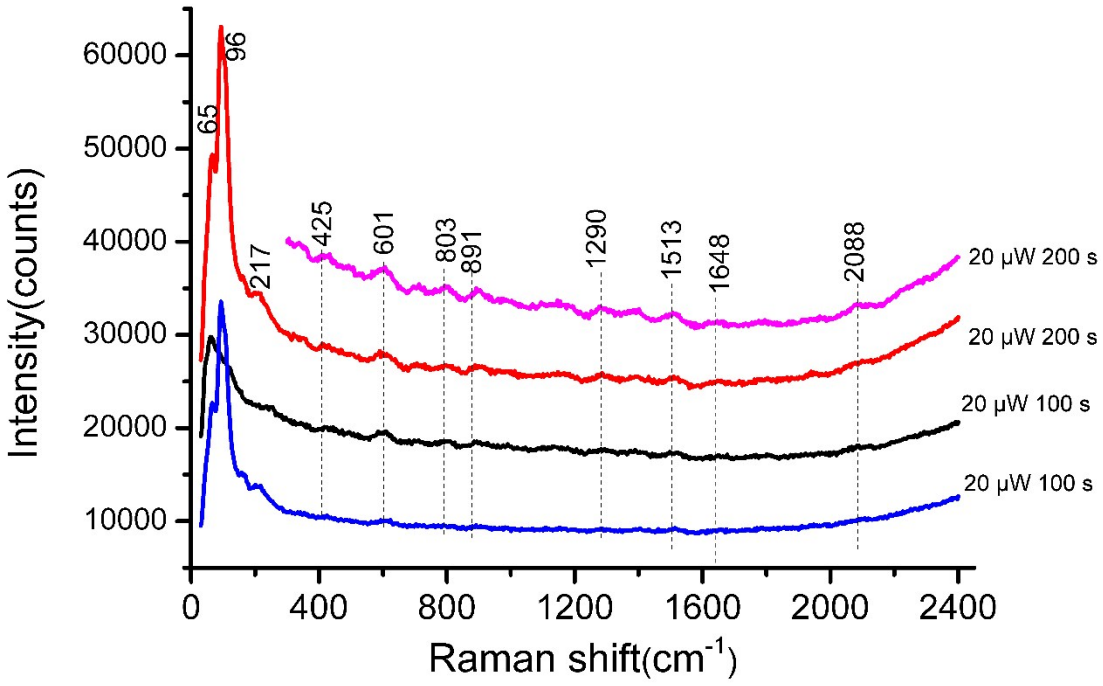

Figure S2. Raman spectra of $MAPbI_3$ single crystals excited by 532 nm laser. The spectra indicated with different colors are measured from different sampling points, demonstrating the reproducibility of the measurements. No peak at around 275 cm^{-1} was observed and the photoluminescence background growth up at the frequency region higher than 2400 cm^{-1}.

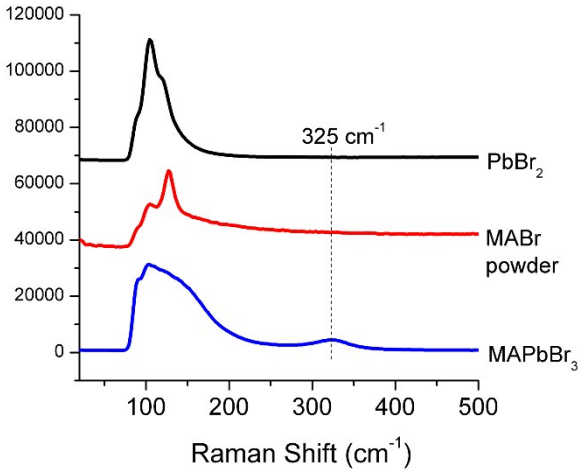

Figure S3. Raman spectra of $MAPbBr_3$ single crystals (blue), MABr powder (red), and $PbBr_2$ (black). No peak were observed for MABr powder and $PbBr_2$, which means that the band 325 cm^{-1} of $MAPbBr_3$ single crystal does come from neither MABr nor $PbBr_2$.

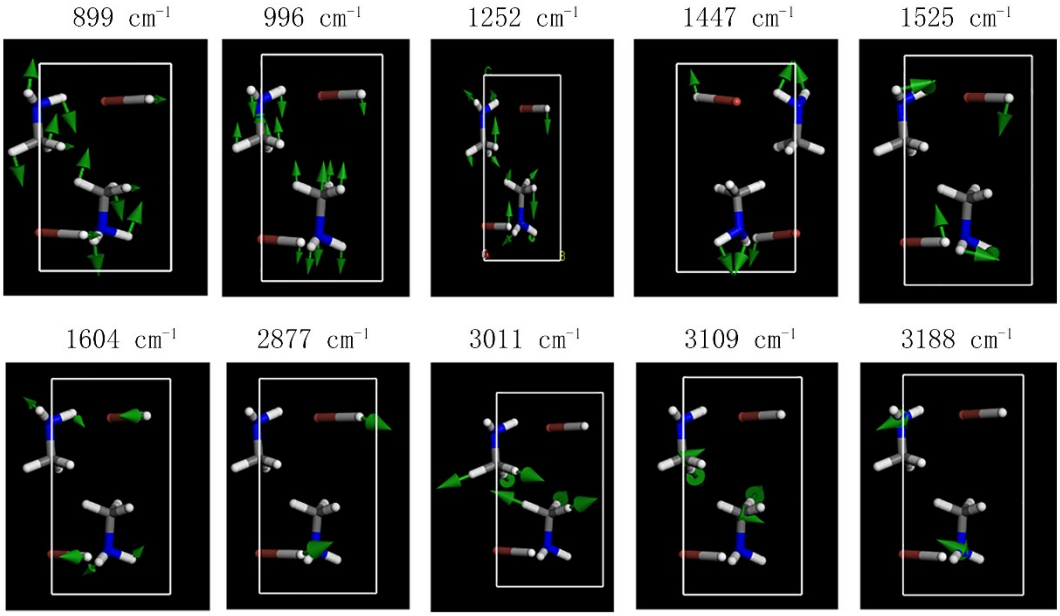

Figure S4.Schematic diagram illustration of CASTEP calculated Raman active modes of MABr solid.

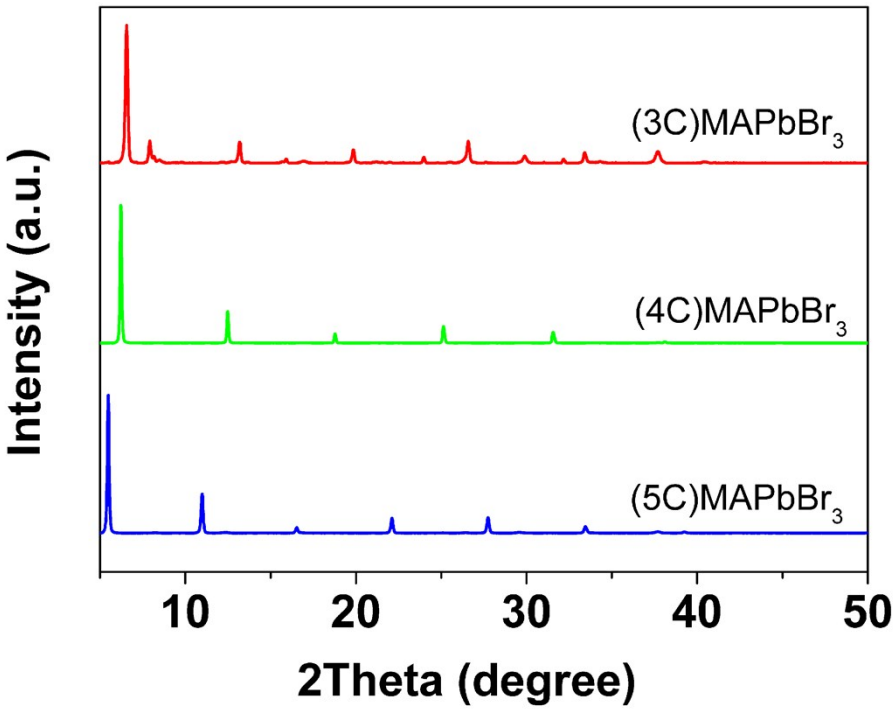

Figure S5 The XRD pattern of 2D perovskite adopt ammonic acids as cation. (3C=β-alanice, 4C=aminobutric acid, 5C=aminovaleric acid）

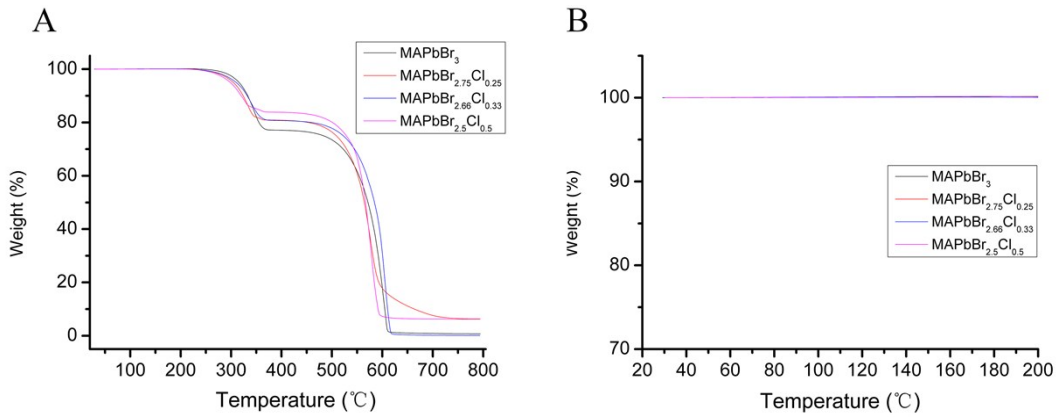

Figure S6 Thermogravimetric analysis (TGA) of $MAPbBr_{3-x}Cl_x$ single crystals in the temperature range of 30 to 800 ℃ (A) and 30 to 800 ℃ (B) carried out in nitrogen flow. These perovskites first undergo HX loss at about 250 ℃, followed by CH_3NH_2 loss at about 345 ℃. There is no weight loss in the temperature range of 30 to 200 ℃, indicating that there is no residual solvent in these crystals . We note that the boiling point of the solvent (DMF) used in the crystal growth is 152.8 ℃.

References

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