Electronic Supplementary Information

Synthesis, Structure, Mobility and Memristors Properties of Tetragonal $CH_3NH_3PbBr_3$ Perovskite Single Crystal

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1. Experimental Section

Preparation of solution for T-$MAPbBr_3$ SC:

Under nitrogen atmosphere, 0.1062 g CH_3NH_3Br (about 1 mmol) was added into 1 mL DMF solution at room temperature, and the solution was colorless and transparent after shaking. Then, 0.3689g $PbBr_2$ (about 1 mmol) was added to the solution to obtain a white turbid solution after full oscillation.

Synthesis of T-$MAPbBr_3$ SC seeds:

The prepared solution in a bottle without a lid was exposed to nitrogen atmosphere. Then, the natural volatilization was made for a period of time until the solvent was completely volatilized and all the crystals were precipitated out. Finally, T-$MAPbBr_3$ SC seeds were obtained.

Growth of T-$MAPbBr_3$ SC:

The prepared solution was placed in an oven and heated according to a certain program (30 to 60 ℃ for 5 minutes, 60 to 80 ℃ for 600 minutes, 80℃ for 240 minutes, and then cooled to 30℃ for 10 minutes). When the temperature rose to 60℃, the crystal seed obtained by natural volatilization was placed in the center of the solution. After the oven heating process was over, the crystal was

taken out and used as a seed for the next growth. The above steps were repeated five times. After five times crystal growth, the crystal size was 12 x 12 x 3 mm.

Fabrication of devices for Cyclic Voltammetry measurements:

Three hairs were fixed on the surface of T-$MAPbBr_3$ SC side by side at a certain distance. Then, the gold was sputtered 20 times with a current of 20 mA under vacuum by sputtering coater.

Fabrication of devices for Hall effect measurements:

The surface of the T-$MAPbBr_3$ SC was partially obscured by A4 paper, exposing four corners of the crystal's upper surface. And the gold was deposited to the crystal by evaporation under a high vacuum.

Characterization: CV (Cyclic Voltammetry) characteristics of the memristor based on T-$MAPbBr_3$ SC at different sweep rates (1, 50, 100, 150, 200, 250 and 300 mV/s) were studied by an electrochemical workstation system (ZAHNER CIMPS-2 pro, Germany). UV-Vis absorption spectra was obtained by UV-Vis NIR spectrometer (PerkinElmer, λ, 750S). The detailed experimental process of diffuse reflection measurements is as follows. Firstly, baseline calibration is performed with standard whiteboard ($BaSO_4$). The sample is then placed on the standard whiteboard for measurements. In the whole experiment, the accuracy of the experiment is guaranteed by the use of integrating sphere. The integrating sphere diagram in Figure S3 has been added to the electronic supplementary information. Single crystal diffraction data was recorded by a Bruker SMART CCD diffractometer with the use of Mo-Kα radiation (λ = 0.71073 Å). Powder X-ray diffraction was performed using PANalyticalX´Pert diffractometer (Cu Kα radiation at λ = 1.54 Å) sampling at 2°/ min, 40 ekV and 100 mA. The morphology and EDS of T-$MAPbBr_3$ SC was characterized by scanning electron microscopy (Thermo Fisher Scientific FIB-SEM GX4, USA). TGA and DSC data were collected by DSC/DTA-TG（NETZSCH, STA449F5-QMS403D, German).

2. Results

Table 1. Crystal data and structure refinement for T-$MAPbBr_3$ SC.

Identification code	T-$MAPbBr_3$ SC
Empirical formula	C1, H6, Br3, N1, Pb1
Temperature/K	298(2) K
Wavelength, A	0.71073
Crystal system	Tetragonal
space group	P4/mmm
a, A	5.9390(5)
b, A	5.9390(5)
c, A	11.8701(11)
alpha, deg	90
beta, deg.	90
gamma , deg	90
Volume, A^3	418.68(6)
Z, Calculated density	1, 3.926
Absorption coefficient, mm^-1	34.368
F(000)	422
Crystal size, mm	0.21 x 0.35 x 0.37
Theta range for data collection , deg.	3.43 to 25.10
Limiting indices	-7<=h<=4, -7<=k<=6, -10<=l<=14
Reflections collected / unique	1989 / 271 [R(int) = 0.1495]
Completeness to theta = 25.10	99.6 %
Absorption correction	Semi-empirical from equivalents
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	271 / 0 / 15
Goodness-of-fit on F^2	1.129
Final R indices [I>2sigma(I)]	R1 = 0.0809, wR2 = 0.1940
R indices (all data)	R1 = 0.1077, wR2 = 0.2107

Largest diff. peak and hole, A^-3	1.776 and -6.663 e.

Table 2. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (A^2 x 10^3) for T-MAPbBr$_3$ SC. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

Atom	x	y	z	U(eq)
Pb(1)	5000	5000	2492(2)	14(1)
Br(2)	5000	0	2527(6)	92(3)
Br(3)	5000	5000	0	92(7)
Br(1)	5000	5000	5000	88(6)

Table 3. Bond lengths [A] and angles [deg] for T-MAPbBr$_3$ SC.

Bond	Bond lengths [A] and angles [deg]
Pb(1)-Br(3)	2.958(2)
Pb(1)-Br(2)#1	2.9698(3)
Pb(1)-Br(2)	2.9698(3)
Pb(1)-Br(2)#2	2.9698(3)
Pb(1)-Br(2)#3	2.9698(3)
Pb(1)-Br(1)	2.977(2)
Br(2)-Pb(1)#4	2.9698(3)
Br(3)-Pb(1)#5	2.958(2)
Br(1)-Pb(1)#7	2.977(2)
Br(3)-Pb(1)-Br(2)#1	90.81(16)
Br(3)-Pb(1)-Br(2)	90.81(16)
Br(2)#1-Pb(1)-Br(2)	89.989(5)
Br(3)-Pb(1)-Br(2)#2	90.81(16)
Br(2)#1-Pb(1)-Br(2)#2	178.4(3)
Br(2)-Pb(1)-Br(2)#2	89.989(5)
Br(3)-Pb(1)-Br(2)#3	90.81(16)
Br(2)#1-Pb(1)-Br(2)#3	89.989(5)
Br(2)-Pb(1)-Br(2)#3	178.4(3)

Br(2)#2-Pb(1)-Br(2)#3	89.989(5)
Br(3)-Pb(1)-Br(1)	180.0
Br(2)#1-Pb(1)-Br(1)	89.19(16)
Br(2)-Pb(1)-Br(1)	89.19(16)
Br(2)#2-Pb(1)-Br(1)	89.19(16)
Br(2)#3-Pb(1)-Br(1)	89.19(16)
Pb(1)#4-Br(2)-Pb(1)	178.4(3)
Pb(1)#5-Br(3)-Pb(1)	180.0
Pb(1)#7-Br(1)-Pb(1)	180.0

Symmetry transformations used to generate equivalent atoms:

#1 -y+1,x,z #2 -y,x,z #3 x,y+1,z

#4 x,y-1,z #5 -x+1,-y+1,-z #6 -x,-y,-z+1

#7 -x+1,-y+1,-z+1

Table 4. Anisotropic displacement parameters (A^2 x 10^3) for T-$MAPbBr_3$ SC.

The anisotropic displacement factor exponent takes the form:

-2 pi^2 [h^2 a*^2 U11 + ... + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
Pb(1)	20(1)	20(1)	3(1)	0	0	0
Br(2)	138(9)	17(4)	120(7)	0	0	0
Br(3)	137(11)	137(11)	0(5)	0	0	0
Br(1)	123(10)	123(10)	19(6)	0	0	0

3.Picture

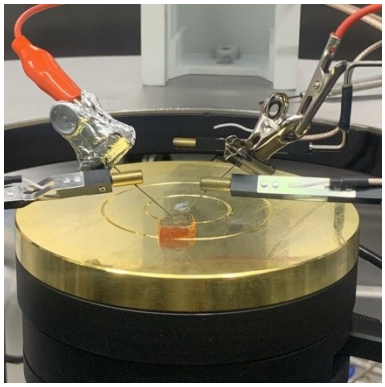

Figure S1 Schematic diagram of test device for memristor device based on T-$MAPbBr_3$ SC

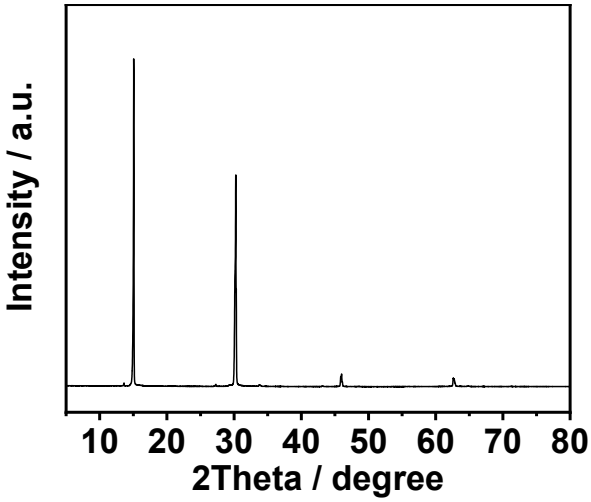

Figure S2 XRD pattern of T-$MAPbBr_3$ SC single crystal

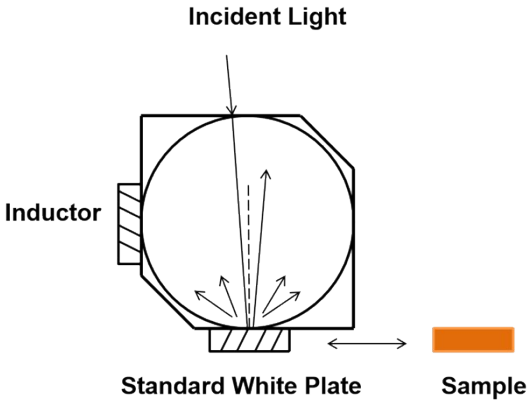

Figure S3 The integrating sphere diagram for diffuse reflection measurements