Supporting Information

$CsPb_2Br_5$ Single Crystals: Synthesis and Characterization

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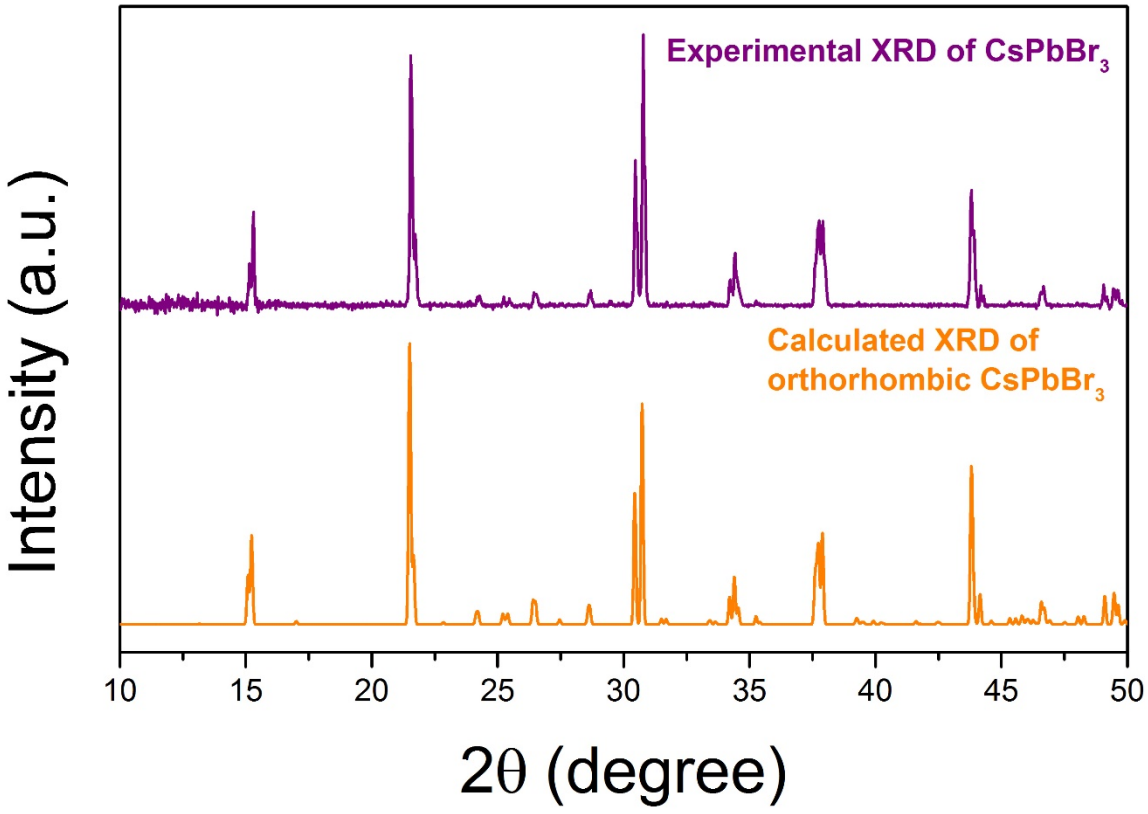

Figure S1: Experimental XRD spectrum of orthorhombic $CsPbBr_3$ from grinded orange crystals and calculated XRD spectrum of orthorhombic $CsPbBr_3$.

Single Crystal XRD

Indexing was performed using APEX2 (Difference Vectors method).[1] Data integration and reduction were performed using SaintPlus 6.01.[2] Absorption correction was performed by analytical method implemented in SADABS.[3] Space groups were determined using XPREP implemented in APEX2.[1] Structure was solved using SHELXS-97 (direct methods) and refined using SHELXL-2014 (full-matrix least-squares on F^2) contained in WinGX.[4] Crystal data and refinement conditions are shown in Table S1.

1. APEX2. Bruker AXS Inc, Madison, Wisconsin, USA, 2014.
2. SAINT. Bruker AXS. Inc, Madison, Wisconsin, USA, 2014.
3. SADABS. G. M. Sheldrick. University of Gottingen, Germany, 2008.
4. SHELXS-97, SHELXL-2014. G. M. Sheldrick, *Acta Cryst.* 2015, C**71**, 3-8; WinGX. L. J. Farrugia, *J. Appl. Cryst.* 2012, **45**, 849-854.

Table S1: Crystal data and structure refinement $CsPb_2Br_5$

Empirical formula	$CsPb_2Br_5$
Formula weight	946.84
Crystal system, space group	Tetragonal, *I*4/*mcm*
Unit cell dimensions	a = 8.4905(4) Å, c = 15.1967(7) Å
Volume	1095.5 (1) $Å^3$
Z, calculated density	4, 5.741 Mg m^{-3}
F(000)	1576
Temperature (K)	296.0(1)
Radiation type	Cu $K\alpha$
Absorption coefficient	105.38 mm^{-1}
Absorption correction	Analytical
Max and min transmission	0.209 and 0.018
Crystal size	0.02 × 0.05 × 0.07 mm
Shape, color	Prism, colorless
θ range for data collection	5.8–66.5°
Limiting indices	$-10 \le h \le 6$, $-10 \le k \le 7$, $-17 \le l \le 17$
Reflection collected / unique / observed with $I > 2\sigma(I)$	3092 / 283 (R_{int} = 0.051) / 281
Completeness to θ_{max} = 66.5°	99.6 %
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	283 / 0 / 16
Final R indices [$I > 2\sigma(I)$]	R_1 = 0.029, wR_2 = 0.071
Final R indices (all data)	R_1 = 0.029, wR_2 = 0.071
Weighting scheme	$[\sigma^2(F_o^2) + (0.0502P)^2 + 0.8505P]^{-1*}$
Extinction coefficient	0.00060(5)
Goodness-of-fit	1.22
Largest diff. peak and hole	1.42 and -3.34 e $Å^{-3}$

$^*P = (F_o^2 + 2F_c^2)/3$

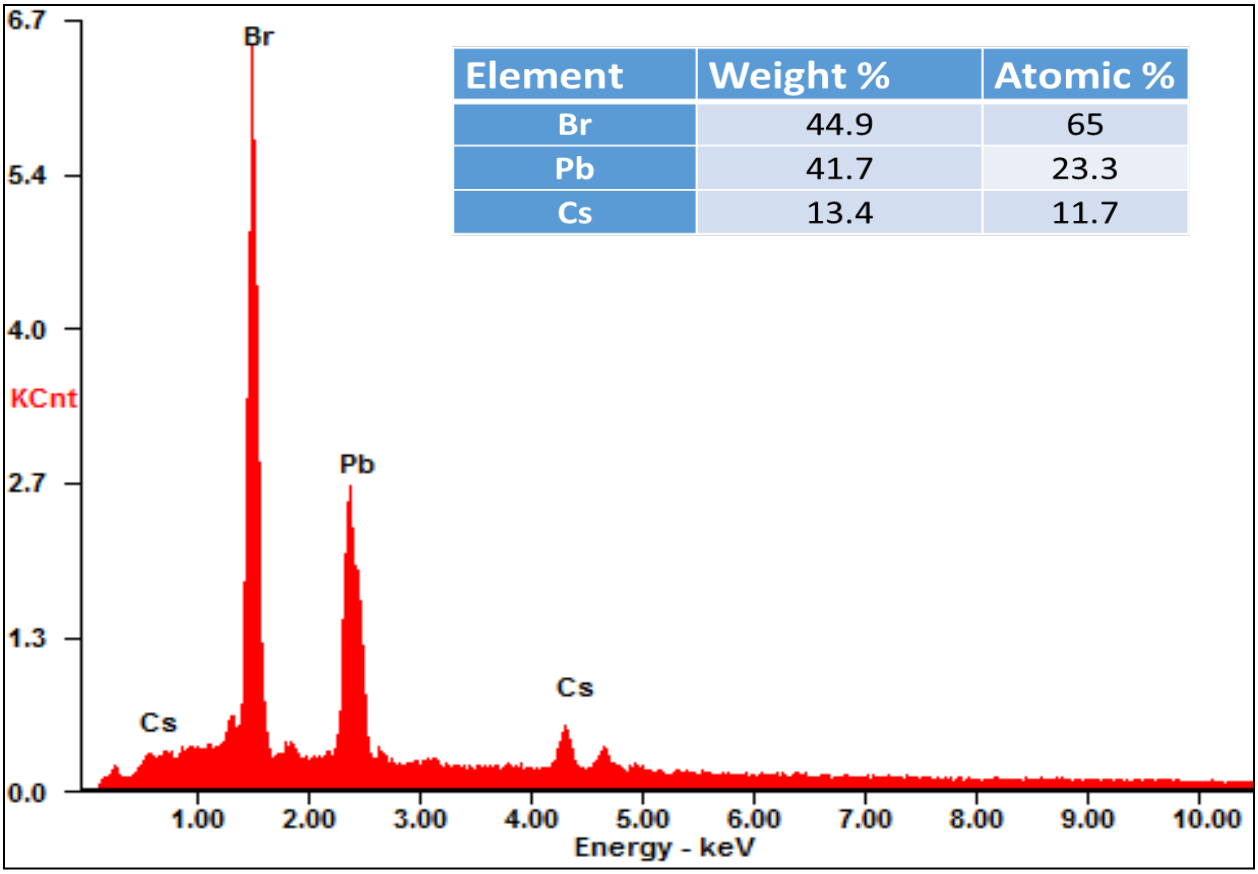

Figure S2: EDS results of the $CsPb_2Br_5$ crystal with atomic ratio values

Density functional theory calculations

Crystal structure optimization and electronic bands calculations were performed using DFT implemented in the PWSCF code of the Quantum ESPRESSO package[1]. The exchange-correlation energy was approximated using the generalized gradient approximation (GGA) proposed by Perdew-Burke-Ernzerhof (PBE). Electron-ion interactions were described by norm-conversing pseudopotentials through explicitly treating electrons for Br ($4s^2$, $4p^5$), Pb ($5d^{10}$, $6s^2$, $6p^2$), and Cs ($5s^2$, $5p^6$, $6s^1$). Single-particle wave functions (charges) were expanded on a plane-wave basis set up to a kinetic energy cutoff of 60 Ry (300 Ry), and Monkhorst-Pack type K-meshes of 4 × 4 × 2 for $CsPb_2Br_5$. The crystal structure of $CsPb_2Br_5$ were fully relaxed until the total force on each atom was less than 0.01 eV/Å. The optical dielectric function of $CsPb_2Br_5$ were calculated using Bethe Salpeter equation (BSE) method implemented in the YAMBO code[2].

References

1. Giannozzi, P.*, et al.* Quantum espresso: A modular and open-source software project for quantum simulations of materials. *J. Phys.: Condens. Matter* **21**, 395502 (2009).

2. Marini, A., Hogan, C., Gruning, M. & Varsano, D. Yambo: An ab initio tool for excited state calculations. *Comput. Phys. Commun.* **180**, 1392-1403 (2009).