

Supplementary Materials

Table S1. Table summarizing peak positions of quasi-single and single Hg_2Br_2 crystals.

Raman, cm^{-1}	Peak Position		Theory [1]	Symmetry		
	Experiment					
	Quasi sing Hg_2Br_2	Singel Hg_2Br_2				
-	-	-	35.5	E_g		
77.7	77.7	77.7	-	-		
94.9	94.9	91	91	E_g		
138	137.7	136	136	A_{1g}		
144.2	143.5	-	-	-		
157.8	157.8	-	-	-		
224.6	222.6	221	221	A_{1g}		

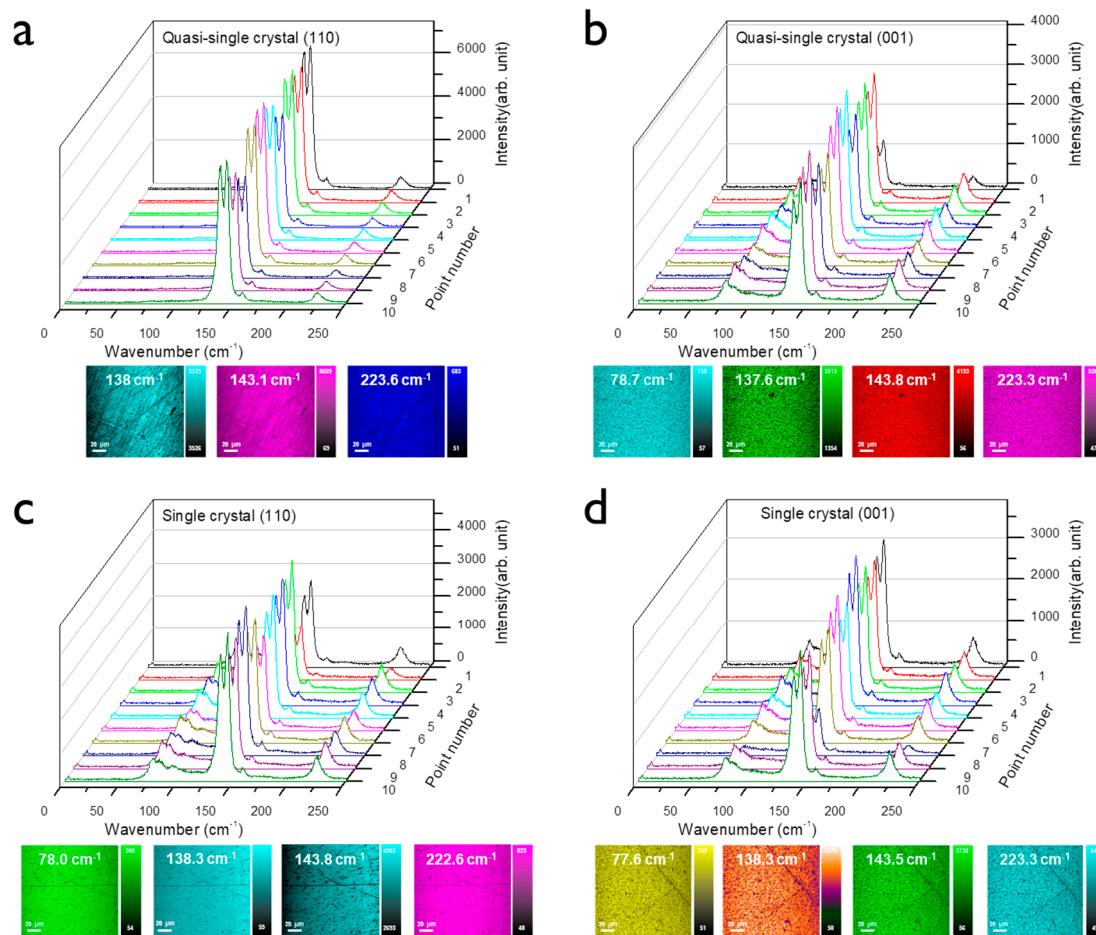


Figure S1. Spectral results extracted from Raman spectroscopy mapping for (110) and (001) planes of quasi-single and single Hg_2Br_2 crystals. The images below show the Raman mapping images for each wavenumber.

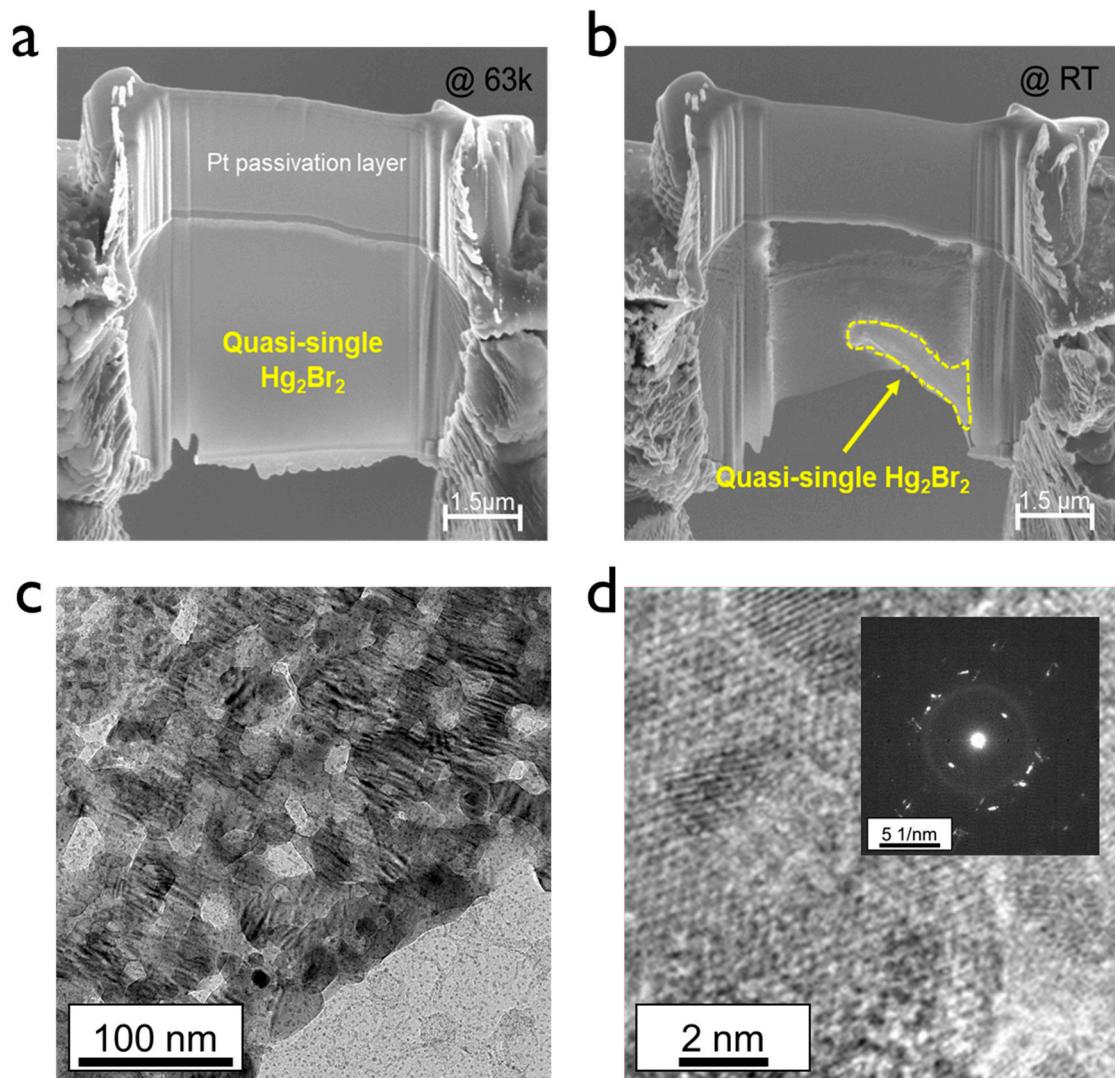


Figure S2. Scanning electron microscopy images of Cryo-FIB sample with quasi-single Hg₂Br₂ at (a) 63K and (b) room temperature; (c) Low magnified and (d) high magnified HRTEM images of the quasi-single Hg₂Br₂ crystal. The inset of (d) shows the corresponding electron diffraction image.

References

1. Roginskii, E.M.; Kvasov, A.A.; Markov, Y.F.; Smirnov, M.B. Lattice dynamics and phonon dispersion in Hg₂Br₂ model ferroelastic crystals. *Tech. Phys. Lett.* **2012**, *38*, 361.



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