



Supplementary Materials

Spectrum	Peak Position			Symmetry
Spectrum	Experiment		- Theory [1]	
	Quasi sing Hg2Br2	Singel Hg ₂ Br ₂	Theory [1]	
Raman, cm⁻¹	-	-	35.5	E_{g}
	77.7	77.7	-	-
	94.9	94.9	91	Eg
	138	137.7	136	A _{1g}
	144.2	143.5	-	-
	157.8	157.8	-	-
	224.6	222.6	221	A _{1g}
Quasi-single crystal (110)		b	Quasi-single crystal (001)	
50 100 150 200 Wavenumber (cm ⁻¹)	2000 2000 2000 0 2000 0 200	0 50 Wave	150 200 enumber (cm ⁻¹)	
138 cm 1 143.1	em ¹ 223.6 cm ¹ ¹⁰	78.7 cm ⁻¹	137.6 cm ⁻¹	143.8 cm ⁻¹ 223.3 (
Single crystal (1	10) 4000 (tion grey/iser	d 0 50	Single crystal (0	201) 30 4 5 4 5 4 5 4 5 4 5 4 5 4 5 4 5 4 5 6 5 4 5 6 5 6 7 6 7 7 7 7 7 7 7 7 7 7 7 7 7
Wavenumber (cm ⁻¹)	001	Wave	enumber (cm ⁻¹)	
) cm ⁻¹ 138.3 cm ⁻¹	145.8 cm ⁻¹ 2 <u>222.6 cm⁻¹</u> 2 <u>22.6 cm⁻¹</u>	77.6 cm ⁻¹	138.3 cm ⁻¹	143.5 cm ⁻¹ 223.3 o

 Table S1. Table summarizing peak positions of quasi-single and single Hg2Br2 crystals.

Figure S1. Spectral results extracted from Raman spectroscopy mapping for (110) and (001) planes of quasi-single and single Hg₂Br₂ 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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