Supplementary material for: "Synthesis and Characterization of $XeAr_2$ under High Pressure"

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TABLE S1: Structural parameters of $XeAr_2$ in the $MgZn_2$-type crystal structure at 10 GPa from DFT calculation. Space group $P6_3/mmc$ (No. 194), a = b = 6.133 Å, c = 9.982 Å.

Atoms	Wyckoff site	x	y	z	Site occupancy	Site Sym
Xe	$4f$	0.33333	0.66667	0.56294	1	$3m$ (100)
Ar1	$2a$	0	0	0	1	-$3m$ (100)
Ar2	$6h$	0.16939	0.33879	0.25000	1	$mm2$ (120)

TABLE S2: Third-order Birch-Murnaghan EoS fittings for $XeAr_2$.

Substance	V_0 ($Å^3$/4.fu)	B_0 (GPa)	B_0'	Source
$XeAr_2$-Exp	613 ± 55	3.7 ± 1.3	4.9 ± 0.3	This work
$XeAr_2$-DFT	600 ± 43	1.6 ± 1.7	5.7 ± 0.7	This work
4*Xe + 8*Ar	672 ± 66	2.8 ± 1.7	5.0 ± 0.4	[1], [2]

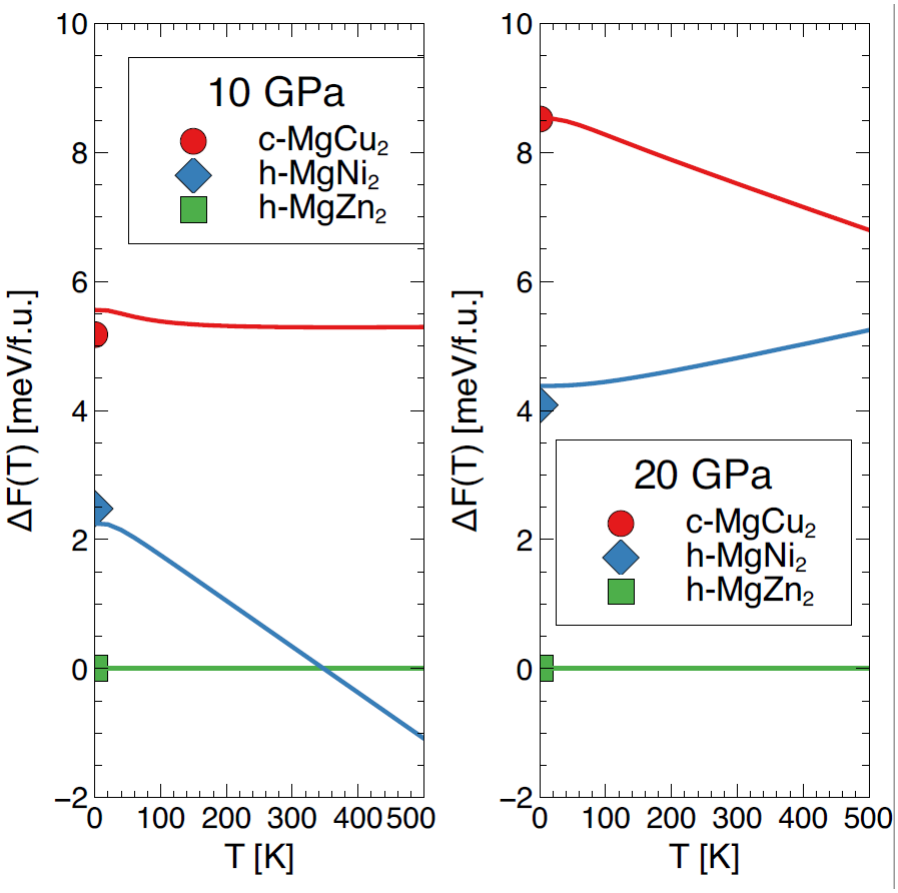

FIG. S1: Relative harmonic vibrational free energies for three structure types of $XeAr_2$, plotted against temperature at P=10 GPa (left) and 20 GPa (right). Big symbols at T = 0 K correspond to relative ground state enthalpies, while lines include zero-point energy (ZPE) and vibrational entropy contributions.

TABLE S3: Optimised volume of $XeAr_2$ in the $MgCu_2$ structure type at P = 20 GPa, using various functionals, divided (from top) into parameterisations of LDA, GGA, and dispersion corrections.

Functional	V [Å^3/f.u.]	V/V_{PBE}	Source
LDA-CA	63.99	0.9274	[3]
LDA-VWN	63.90	0.9261	[4]
PW91	68.79	0.9970	[5]
PBE	69.00	1.0000	[6]
revPBE	71.10	1.0304	[7]
RPBE	71.88	1.0417	[8]
PBEsol	65.88	0.9548	[9]
AM05	67.02	0.9713	[10]
PBE+D2	69.84	1.0122	[11]
PBE+D3	68.76	0.9965	[12]
PBE+D3(BJ)	67.59	0.9796	[13]
PBE+TS	68.85	0.9978	[14]
PBE+TS(HI)	68.76	0.9965	[15]
PBE+MBD	68.43	0.9917	[16]
PBE+dDsC	67.05	0.9717	[17]
PBE+ulg	68.07	0.9865	[18]

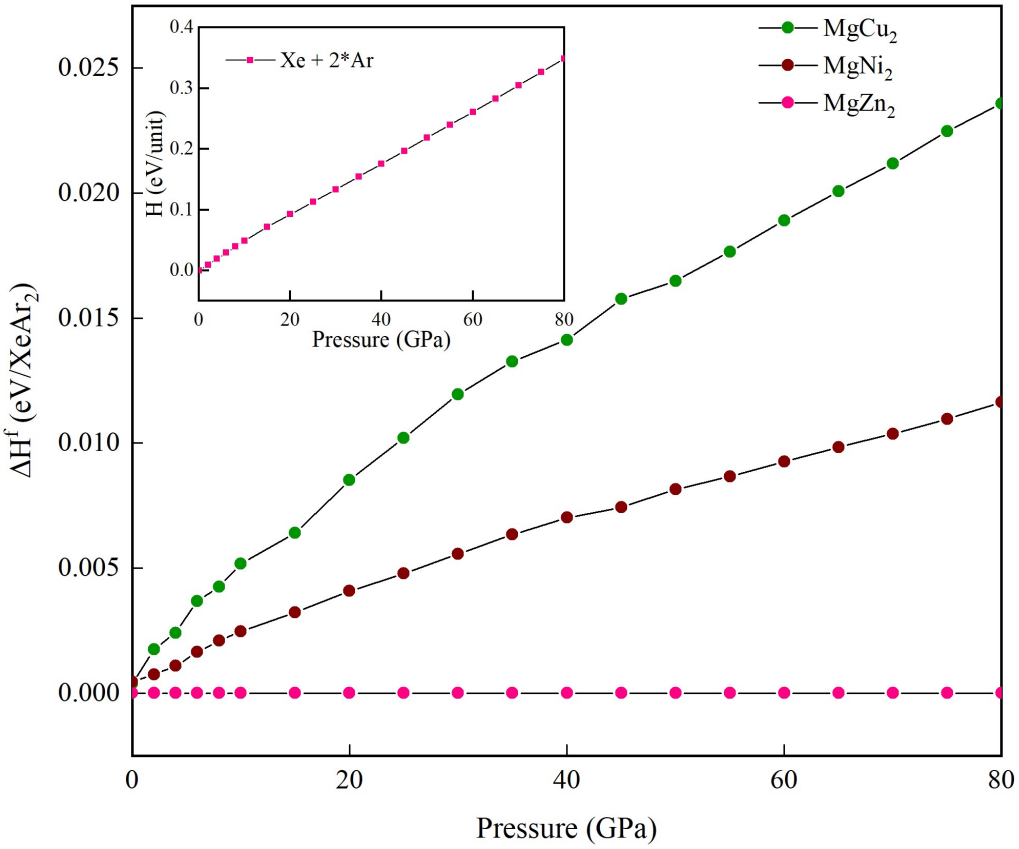

FIG. S2: Ground state formation enthalpies of different $XeAr_2$ structures, relative to the h-$MgZn_2$ structure type, as function of pressure, from DFT-PBE calculations. Note the strongly positive relative enthalpy of the constituents Xe + 2*Ar.

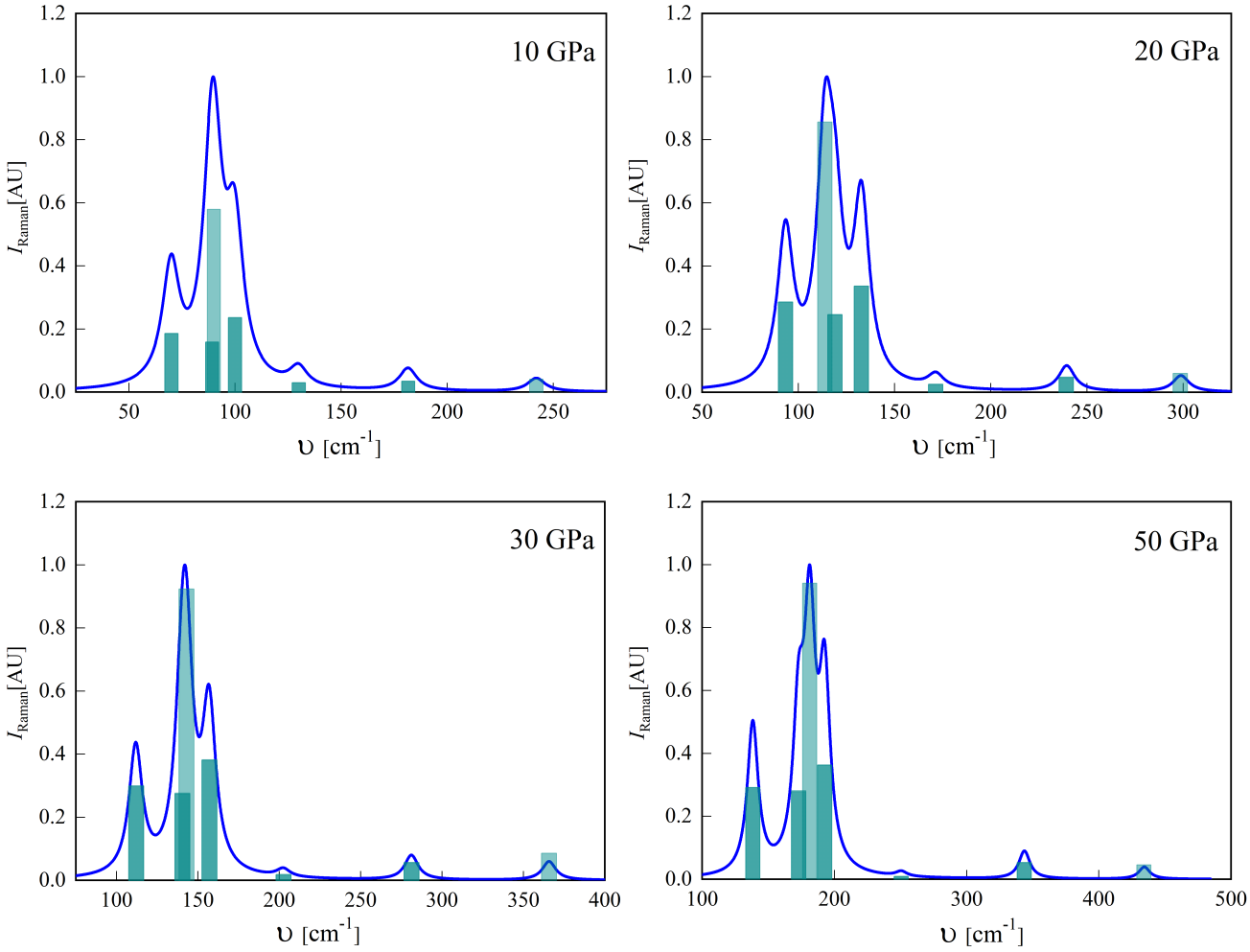

FIG. S3: Simulated Raman spectra of $XeAr_2$ at 10 GPa, 20 GPa, 30 GPa and 50 GPa, respectively, in the h-$MgZn_2$ structure type. We have used a fixed FWHM of 10 cm^{-1} for all modes.

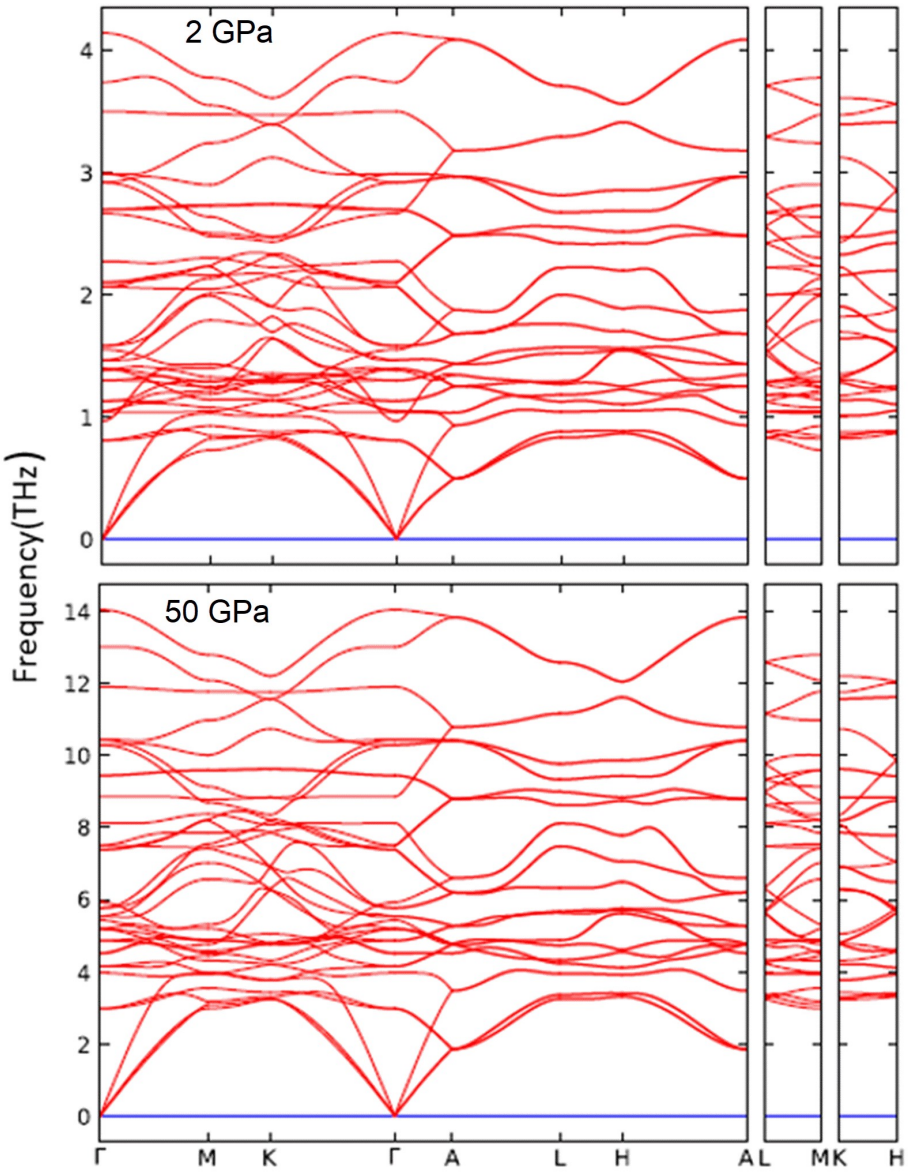

FIG. S4: DFT-PBE phonon dispersions of $XeAr_2$ at 2 GPa (top) and 50 GPa (lower).

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