

Supporting Information

Table S1 Least-Squares Correlation Matrix (p_{1-7} are Geometric Parameters, u_{1-5} Vibrational Amplitudes, k_1 and k_2 Scale Factors)

p_1	p_2	p_3	p_4	p_5	p_6	p_7	u_1	u_2	u_3	u_4	u_5	k_1	k_2
100	49	-8	18	0	0	-8	-6	20	40	-60	17	-16	0
	100	-32	-25	1	3	0	-42	-18	3	-37	-28	-55	-27
		100	51	0	0	0	-4	-57	-61	17	-22	20	8
			100	-1	2	0	7	-35	-10	0	-20	18	17
				100	0	0	0	0	0	0	-2	0	0
					100	0	6	4	5	5	5	13	6
						100	0	-2	-3	20	-2	2	0
							100	43	53	16	45	77	74
								100	82	-15	83	31	28
									100	-27	59	25	39
										100	-6	30	13
											100	41	34
												100	59
													100

Table S2 (a) Cartesian Coordinates Obtained for Gallaborane in the Gas Phase by Electron Diffraction

	x	y	z
Ga1	0	0	0
B2	2.1972	0	0
H3	1.4598	1.0531	0
H4	1.4598	-1.0531	0
H5	-0.6553	0	1.4106
H6	-0.6553	0	-1.4106
H7	2.785	0	1.0341
H8	2.785	0	-1.0341

(b) Atomic Coordinates ($\times 10^4$) for Crystalline Gallaborane at 110 K

	x	y	z
Ga(1)	5954	3849	13089
Ga(2)	7500	6868	10000
B(1)	6400	5664	11524
B(2)	7500	3313	15000
H(1A)	4831	4560	13419
H(1B)	6107	2496	12488
H(2)	6295	7346	9258
H(11)	6392	6856	11718
H(12)	5412	5198	11411
H(13)	7154	5297	12360
H(14)	6968	5234	10818
H(21)	6611	2840	14895
H(22)	7568	4126	14196

Table S3 Computed Parameters for $\text{H}_2\text{Ga}(\mu\text{-H})_2\text{BH}_2$

	$r(\text{Ga}\cdots\text{B})$	$r(\text{Ga}-\text{H}_\text{b})$	$r(\text{B}-\text{H}_\text{b})$	$r(\text{Ga}-\text{H}_\text{a})$	$r(\text{B}-\text{H}_\text{a})$	$\angle\text{H}_\text{a}\text{GaH}_\text{a}$	$\angle\text{H}_\text{b}\text{BH}_\text{a}$	$\angle\text{H}_\text{b}\text{GaH}_\text{b}$	$\angle\text{H}_\text{a}\text{BH}_\text{b}$	
HF	6-311G(d)	2.239	1.788	1.303	1.557	1.192	129.7	121.3	71.1	105.9
	6-311G(d,p)	2.230	1.781	1.302	1.555	1.191	129.4	121.1	71.4	105.9
	6-311g(2d,2p)	2.230	1.782	1.300	1.557	1.188	129.5	121.0	71.3	106.1
	6-311+G(d)	2.238	1.787	1.303	1.558	1.192	129.5	121.2	71.2	106.0
	6-311+G(d,p)	2.230	1.781	1.302	1.555	1.191	129.4	121.1	71.4	106.0
MP2(FC)	6-311+g(2d,2p)	2.230	1.782	1.300	1.557	1.188	129.5	121.0	71.3	106.1
	6-311++g(2d,2p)	2.230	1.782	1.300	1.557	1.188	129.5	121.0	71.3	106.1
	6-311G(d)	2.222	1.805	1.287	1.562	1.195	130.7	121.1	70.8	108.6
	6-311G(2d,2p)	2.220	1.793	1.281	1.562	1.189	130.1	120.9	70.5	107.7
	6-311G(2df,2pd)	2.215	1.791	1.280	1.561	1.193	130.2	120.9	70.6	107.9
MP2(full)	6-311+G(2d,2p)	2.220	1.793	1.281	1.562	1.189	130.1	120.9	70.5	107.7
	6-311+G(2df,2pd)	2.215	1.791	1.280	1.561	1.190	130.1	120.9	70.6	107.9
	6-311++G(2d,2p)	2.220	1.793	1.281	1.562	1.189	130.1	120.9	70.5	107.7
	6-311G(d)	2.195	1.783	1.288	1.549	1.194	130.9	121.1	71.9	108.7
	6-311g(d,p)	2.189	1.761	1.286	1.535	1.192	130.4	121.1	72.0	107.1
MP3(FC)	6-311g+(d)	2.195	1.783	1.288	1.549	1.194	130.9	121.1	71.9	108.7
	6-311g+(d,p)	2.189	1.761	1.286	1.535	1.192	130.4	121.1	72.0	107.1
	6-311G(d)	2.220	1.807	1.291	1.571	1.197	130.8	120.9	71.1	108.9
	6-311G(d,p)	2.222	1.790	1.288	1.560	1.194	130.5	120.9	70.8	107.3
	6-311+G(d)	2.220	1.807	1.291	1.571	1.197	130.8	120.9	71.1	108.9
MP3(full)	6-311+G(d,p)	2.222	1.790	1.288	1.560	1.194	130.5	120.9	70.8	107.3
	6-311+G(d)	2.222	1.790	1.288	1.560	1.194	130.5	120.9	70.8	107.3
	6-311G(d)	2.207	1.797	1.290	1.563	1.197	130.9	122.0	71.5	109.0
	6-311G(d)	2.225	1.809	1.294	1.574	1.199	130.8	120.9	71.1	108.8
	6-311G(d,p)	2.225	1.790	1.291	1.562	1.195	130.5	121.0	70.9	107.1
MP4(SDQ,FC)	6-311G(d,p)	2.225	1.790	1.291	1.562	1.195	130.5	121.0	70.9	107.1
	6-311+G(d)	2.225	1.809	1.294	1.574	1.199	130.8	120.9	71.1	108.8
	6-311+G(d,p)	2.225	1.790	1.291	1.562	1.195	130.5	121.0	70.9	107.1
	6-311+G(d)	2.225	1.809	1.294	1.574	1.199	130.8	120.9	71.1	108.8
	6-311G(d)	2.225	1.790	1.291	1.562	1.195	130.5	121.0	70.9	107.1
B3PW91	6-311G(d,p)	2.195	1.778	1.291	1.548	1.194	130.6	121.4	71.9	108.2
	6-311G(2d,2p)	2.192	1.777	1.289	1.549	1.193	130.1	121.1	72.0	108.2
	6-311G(2df,2pd)	2.190	1.776	1.288	1.548	1.193	130.0	121.0	72.0	108.3
	6-311+G(d)	2.202	1.783	1.293	1.551	1.196	130.6	121.4	71.9	108.2
	6-311+G(d,p)	2.194	1.778	1.291	1.549	1.194	130.3	121.1	72.1	108.2
GED	6-311+G(2d,2p)	2.192	1.777	1.289	1.549	1.193	130.1	121.1	72.0	108.3
	6-311+G(2df,2pd)	2.190	1.776	1.288	1.548	1.193	130.0	121.0	72.0	108.3
	6-311++G(2d,2p)	2.192	1.777	1.289	1.549	1.193	130.1	121.1	72.0	108.3
	6-311++G(2d,2p)	2.192	1.777	1.289	1.549	1.193	130.1	121.1	72.0	108.3
	6-311++G(2d,2p)	2.197(4)	1.800(6)	1.286(7)	1.555(6)	1.190(7)	130.5(9)	120.8(9)	71.6(4)	110.0(5)