Electronic Supplementary Information for:

The Structure of Monoclinic $Na_2B_{10}H_{10}$: A Combined Diffraction, Spectroscopy, and Theoretical Approach

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How to view phonon animations using the V_Sim software*

The **Na2B10H10_phonons.xyz** and **Na2B10D10_phonons.xyz** files contain the information needed to view the animated (gamma-point) phonon normal modes from the DFT-optimized 0 K $Na_2{}^{11}B_{10}H_{10}$ and $Na_2{}^{11}B_{10}D_{10}$ structures, respectively, following the steps below:

- Get the V_Sim software (It is free, and there is no need to install).
- Go to the following webpage and download the Win32 binaries:
  http://www-drfmc.cea.fr/L_Sim/V_Sim/download.html
- Unzip the zip file to wherever you want to put the software.

- Click "~V_Sim\bin\V_sim.exe" to start the V_Sim program, then open the **Na2B10H10_phonons.xyz** or **Na2B10D10_phonons.xyz** file to view the phonon animations.
- To build bonds in the structure, check the box on the left side of the "Pairs" button, and click the "Pairs" button.
- Highlight a pair and click the "Auto set" button to allow bonding.
- Adjust the "Link parameters," if desired.
- Adjust the element color, radius, etc. on the "Elements" tab, if desired.
- Go to the "Phonons" tab, highlight a phonon mode, and click the "Play" button to view a phonon animation.

* N.B., the use of this software does not imply its recommendation or endorsement by NIST.

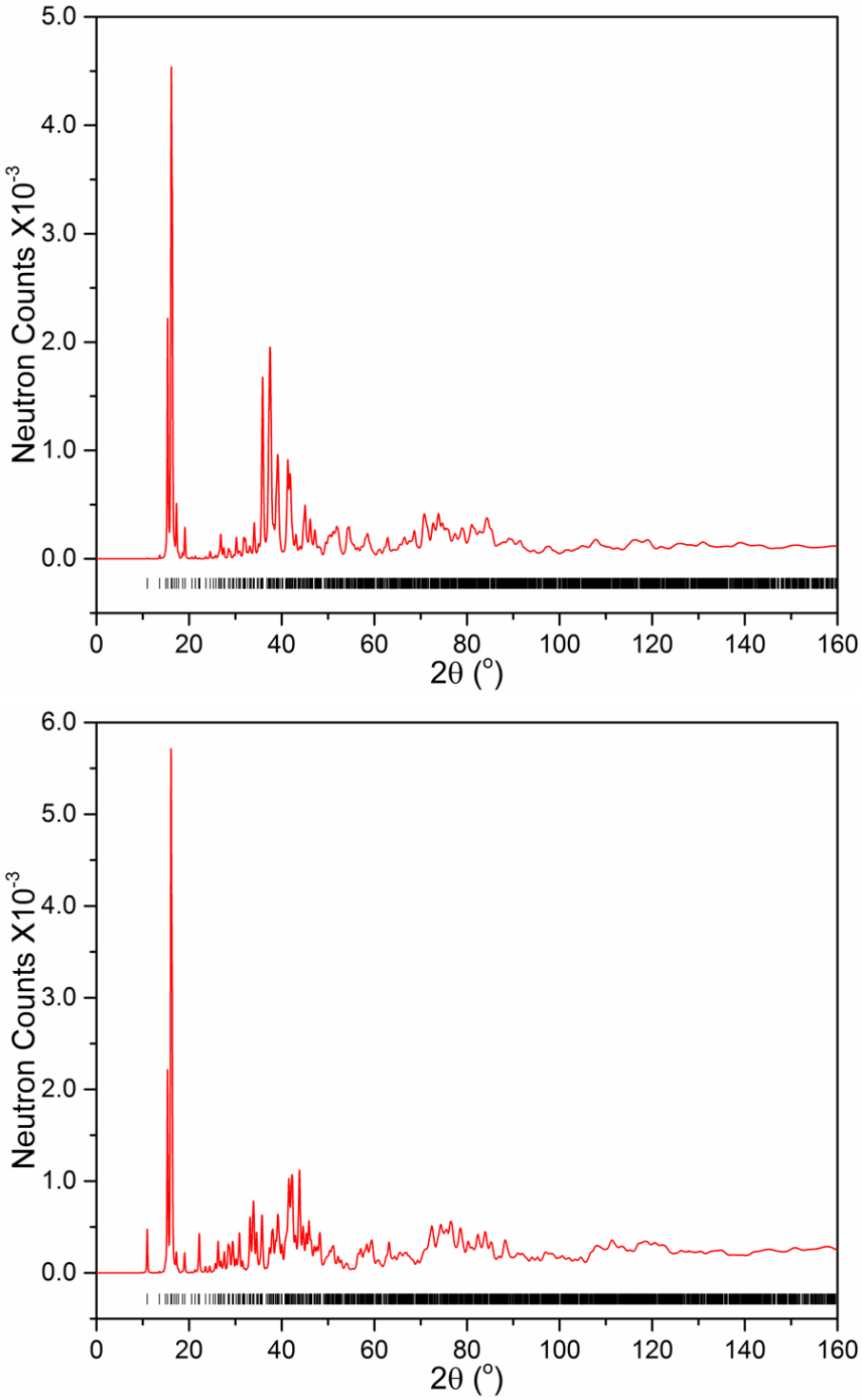

Fig. S1 Comparison of the simulated $Na_2{}^{11}B_{10}D_{10}$ NPD patterns (assuming λ=1.5398 Å) for the (top) modified 2.5 K and (bottom) published [from K. Hofmann and B. Albert, *Z. Kristallogr.*, 2005, **220**, 142-146] 100 K structures, clearly indicating the broad disagreement between the two structure models and confirming the substantial sensitivity of NPD to the particular structural details.

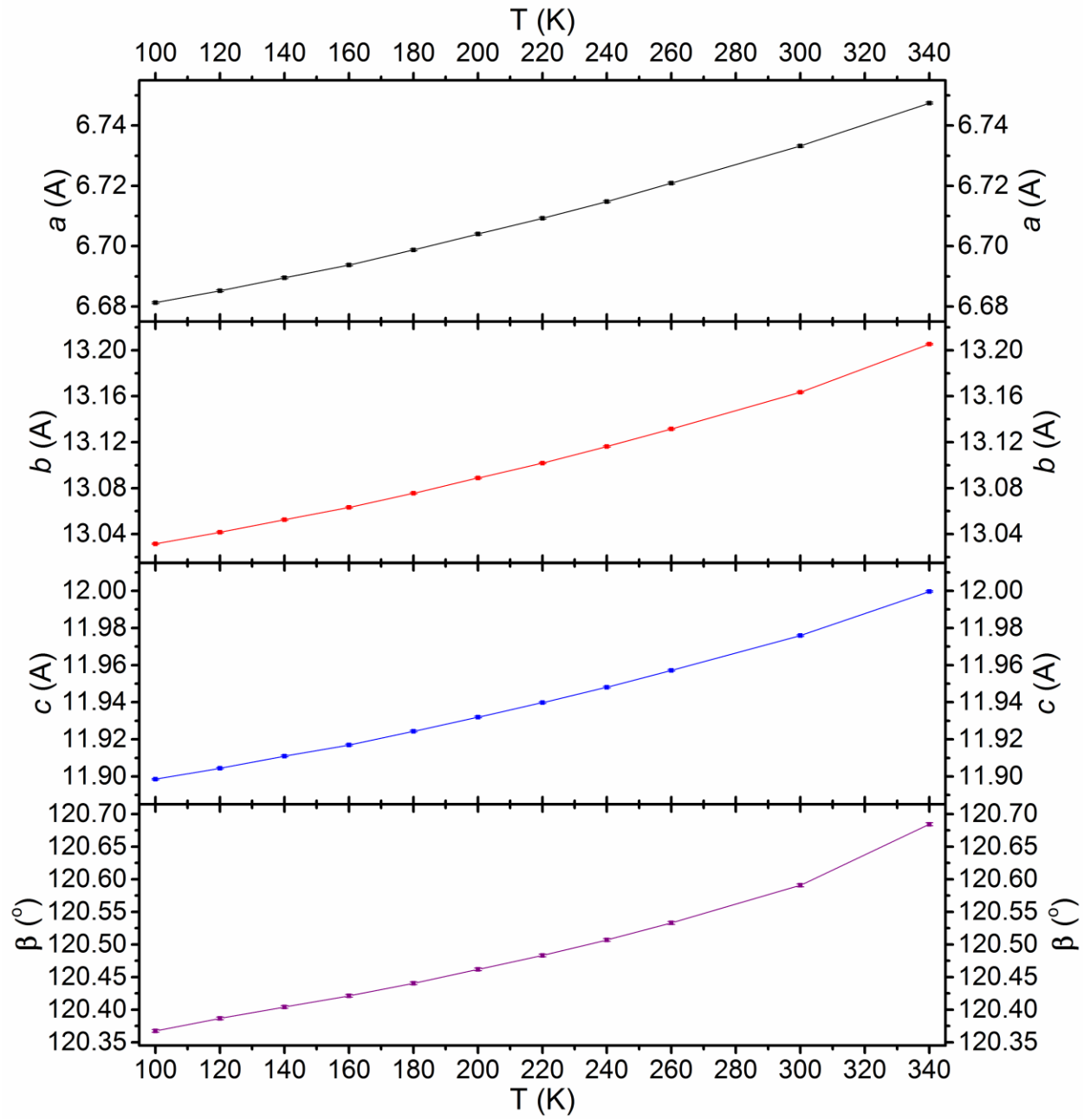

Fig. S2 Plot of the monoclinic lattice parameters versus temperature for $Na_2B_{10}H_{10}$ based on synchrotron XRPD data. Standard uncertainties are smaller than the symbol size.

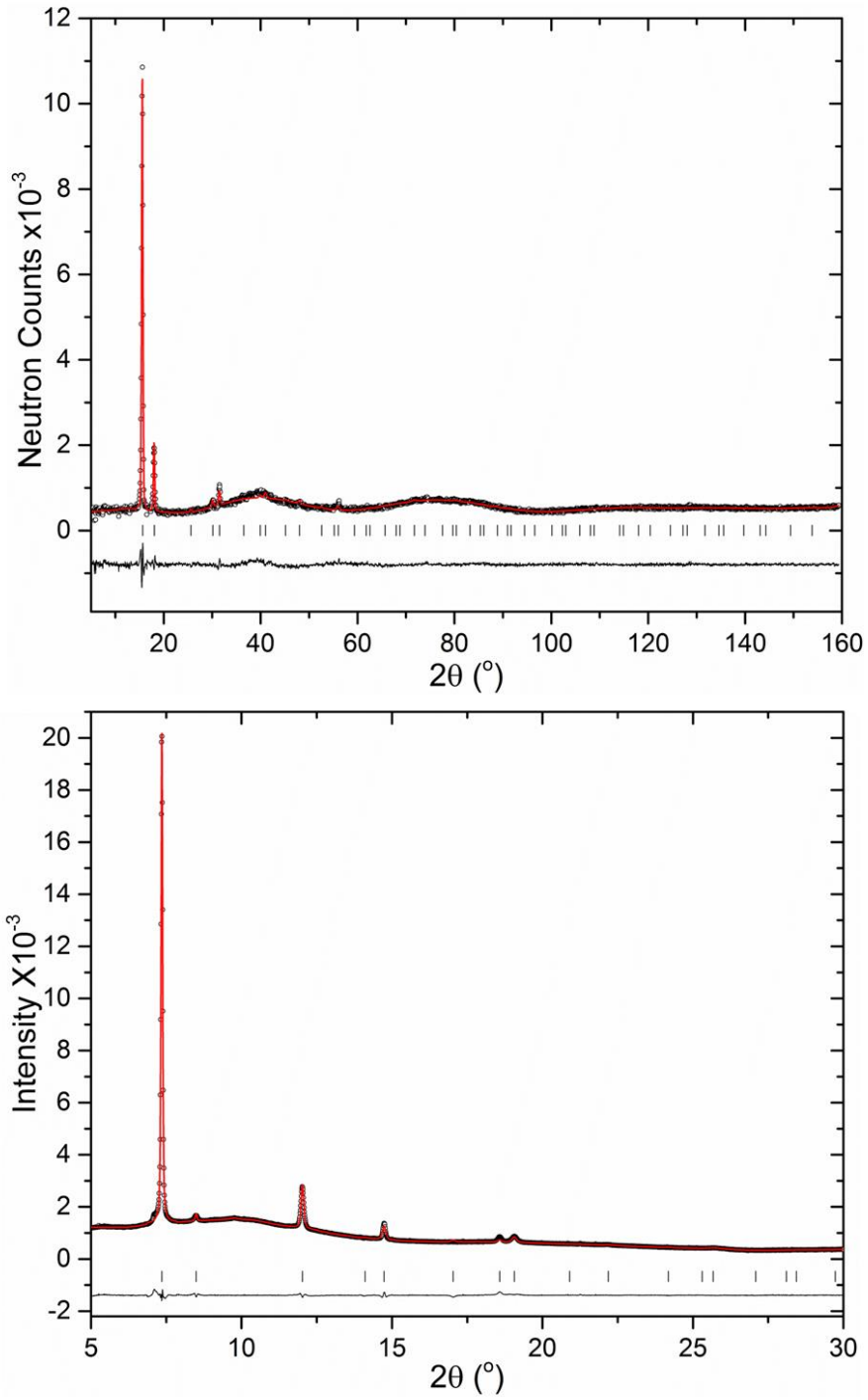

Fig. S3 Experimental (circles), fitted (line), and difference (line below observed and calculated patterns) profiles at 410 K for the high-temperature cubic structure [space group *Fm*-3*m*; from T. J. Udovic et al., *Adv. Mater.*, 2014, **26**, 7622-7626] of (top) $Na_2{}^{11}B_{10}D_{10}$ (NPD, λ=1.5398(2) Å, *a*=9.8397(5) Å, R_{wp}=0.0491, R_p=0.0412, χ^2=1.403) and (bottom) $Na_2B_{10}H_{10}$ (synchrotron XRPD, λ=0.7296(1) Å, *a*=9.8517(6), R_p=0.136, R_{wp}=0.0924, χ^2=0.482). Vertical bars indicate the calculated positions of the Bragg peaks. This structure is the same as that reported by Udovic et al. except for small changes in the Na site occupancies. (See Tables S5 and S6.) Standard uncertainties are commensurate with the scatter in the data.

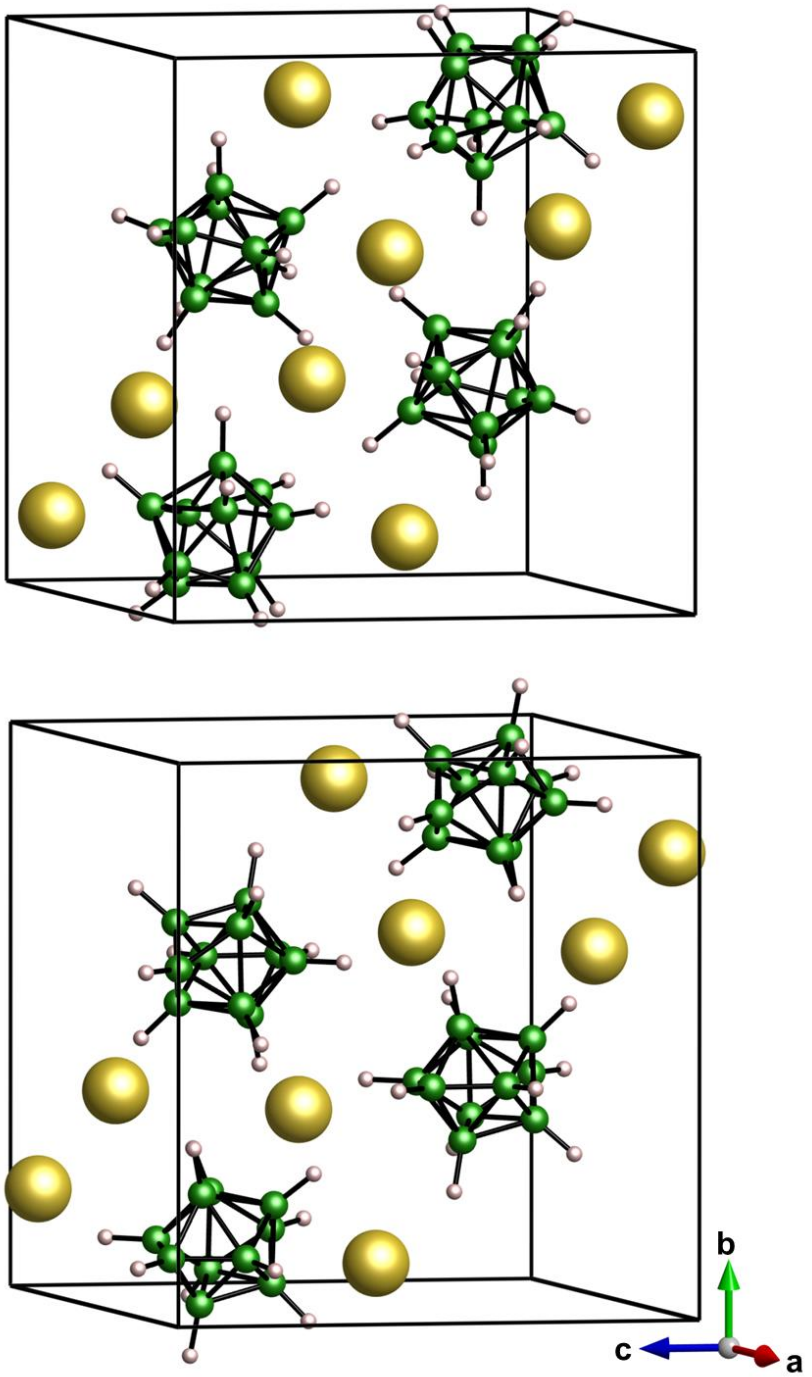

Fig. S4 Comparison of the published monoclinic $Na_2B_{10}H_{10}$ crystal structure [from K. Hofmann and B. Albert, *Z. Kristallogr.*, 2005, **220**, 142-146] (top) before and (bottom) after energy-optimization. Yellow, green, and white spheres denote Na, B, and D atoms, respectively. Although the energy-optimized structure is the nearest stable configuration to the published structure, there are significant differences between the two structures with respect to anion distortions and orientations and cation positions.

Table S1. The refined $Na_2{}^{11}B_{10}D_{10}$ structural parameters at 2.5 K associated with the monoclinic space group $P2_1/c$ (No.14) derived from NPD data.

$Na_2{}^{11}B_{10}D_{10}$		**T = 2.5 K**			**NPD Cu(311)/λ=1.5398(2) Å**	
a = 6.6535(5) Å; b= 12.964(1) Å; c = 11.8510(9) Å; γ=120.203(4) °; V = 883.4(1) Å^3; Z = 4						
Atom	Site	x	y	z	U_{iso} (Å^2)	Occ.
Na1	4*e*	0.239(3)	0.394(2)	0.492(2)	0.030(3)	1
Na2	4*e*	0.216(3)	0.630(2)	0.015(2)	0.030(3)	1
B1	4*e*	0.5610(5)	0.5103(2)	0.7580(3)	0.0144(2)	1
B2	4*e*	0.4361(5)	0.5967(2)	0.6170(2)	0.0144(2)	1
B3	4*e*	0.5465(5)	0.7198(2)	0.6933(2)	0.0144(2)	1
B4	4*e*	0.7372(4)	0.6079(2)	0.7438(3)	0.0144(2)	1
B5	4*e*	0.2897(4)	0.6692(2)	0.6872(2)	0.0144(2)	1
B6	4*e*	0.4144(5)	0.5822(2)	0.8284(2)	0.0144(2)	1
B7	4*e*	0.7204(4)	0.5980(2)	0.8928(2)	0.0144(2)	1
B8	4*e*	0.5293(5)	0.7098(2)	0.8431(2)	0.0144(2)	1
B9	4*e*	0.2726(4)	0.5390(2)	0.6725(3)	0.0144(2)	1
B10	4*e*	0.7846(5)	0.7106(2)	0.8439(3)	0.0144(2)	1
D1	4*e*	0.6218(6)	0.4221(2)	0.7663(4)	0.0233(4)	1
D2	4*e*	0.3890(6)	0.5842(3)	0.5056(2)	0.0233(4)	1
D3	4*e*	0.5432(7)	0.7922(2)	0.6292(3)	0.0233(4)	1
D4	4*e*	0.8890(5)	0.5818(3)	0.7233(4)	0.0233(4)	1
D5	4*e*	0.1176(5)	0.7224(3)	0.6406(4)	0.0233(4)	1
D6	4*e*	0.3466(6)	0.5522(3)	0.8991(3)	0.0233(4)	1
D7	4*e*	0.8515(6)	0.5629(3)	0.9997(2)	0.0233(4)	1
D8	4*e*	0.4820(7)	0.7709(2)	0.9024(3)	0.0233(4)	1
D9	4*e*	0.1078(5)	0.4824(3)	0.6226(3)	0.0233(4)	1
D10	4*e*	0.9491(6)	0.7675(3)	0.8946(3)	0.0233(4)	1

Table S2. The refined $Na_2{}^{11}B_{10}D_{10}$ structural parameters at 295 K associated with the monoclinic space group $P2_1/c$ (No.14) derived from NPD data.

$Na_2{}^{11}B_{10}D_{10}$		**T = 295 K**			**NPD Cu(311)/λ=1.5398(2) Å**	
a = 6.7137(7) Å; b= 13.124(1) Å; c = 11.940(1) Å; γ= 120.524(6) °; V = 906.2(2) Å^3; Z = 4						
Atom	Site	x	y	z	U_{iso} (Å^2)	Occ.
Na1	4*e*	0.217(6)	0.399(3)	0.484(4)	0.094(7)	1
Na2	4*e*	0.224(6)	0.625(3)	-0.012(4)	0.094(7)	1
B1	4*e*	0.5645(8)	0.5121(2)	0.7598(4)	0.0287(3)	1
B2	4*e*	0.4351(7)	0.5967(3)	0.6188(4)	0.0287(3)	1
B3	4*e*	0.5424(8)	0.7190(2)	0.6937(4)	0.0287(3)	1
B4	4*e*	0.7352(6)	0.6096(3)	0.7442(5)	0.0287(3)	1
B5	4*e*	0.2902(6)	0.6675(3)	0.6890(5)	0.0287(3)	1
B6	4*e*	0.4195(7)	0.5822(3)	0.8303(4)	0.0287(3)	1
B7	4*e*	0.7227(6)	0.5996(3)	0.8933(4)	0.0287(3)	1
B8	4*e*	0.5296(7)	0.7089(3)	0.8435(4)	0.0287(3)	1
B9	4*e*	0.2769(7)	0.5388(3)	0.6754(4)	0.0287(3)	1
B10	4*e*	0.7815(7)	0.7112(3)	0.8433(5)	0.0287(3)	1
D1	4*e*	0.627(1)	0.4253(2)	0.7685(6)	0.0489(7)	1
D2	4*e*	0.386(1)	0.5842(4)	0.5077(4)	0.0489(7)	1
D3	4*e*	0.536(1)	0.7905(3)	0.6291(5)	0.0489(7)	1
D4	4*e*	0.8853(8)	0.5846(4)	0.7232(6)	0.0489(7)	1
D5	4*e*	0.1177(7)	0.7190(4)	0.6428(6)	0.0489(7)	1
D6	4*e*	0.355(1)	0.5522(4)	0.9015(5)	0.0489(7)	1
D7	4*e*	0.8558(8)	0.5656(4)	0.9998(4)	0.0489(7)	1
D8	4*e*	0.482(1)	0.7689(3)	0.9026(6)	0.0489(7)	1
D9	4*e*	0.1145(9)	0.4820(4)	0.6267(5)	0.0489(7)	1
D10	4*e*	0.9435(8)	0.7683(4)	0.8928(6)	0.0489(7)	1

Table S3. The refined $Na_2B_{10}H_{10}$ structural parameters at 100 K associated with the monoclinic space group $P2_1/c$ (No. 14) derived from XRPD data (R_p=0.0811, R_{wp}=0.0732, χ^2=0.379).

$Na_2B_{10}H_{10}$		**T = 100 K**			**XRPD**	**λ=0.7296(1) Å**
a = 6.6812(3) Å;	*b*= 13.0314(6) Å;	*c* = 11.8984(4) Å;	γ=120.368(2) °;	*V* = 893.81(6) Å^3;	Z = 4	
Atom	Site	*x*	*y*	*z*	U_{iso} (Å^2)	Occ.
Na1	4*e*	0.229(1)	0.3877(7)	0.4845(7)	0.028(1)	1
Na2	4*e*	0.201(1)	0.6296(8)	0.0064(7)	0.028(1)	1
B1	4*e*	0.561(1)	0.5124(4)	0.7595(7)	0.008(1)	1
B2	4*e*	0.436(1)	0.5961(5)	0.6161(5)	0.008(1)	1
B3	4*e*	0.540(1)	0.7193(4)	0.6887(6)	0.008(1)	1
B4	4*e*	0.735(1)	0.6101(5)	0.7438(6)	0.008(1)	1
B5	4*e*	0.2825(9)	0.6678(5)	0.6828(6)	0.008(1)	1
B6	4*e*	0.409(1)	0.5846(5)	0.8273(6)	0.008(1)	1
B7	4*e*	0.714(1)	0.6025(5)	0.8919(5)	0.008(1)	1
B8	4*e*	0.519(1)	0.7111(5)	0.8388(7)	0.008(1)	1
B9	4*e*	0.271(1)	0.5391(5)	0.6724(7)	0.008(1)	1
B10	4*e*	0.774(1)	0.7141(5)	0.8401(7)	0.008(1)	1
H1	4*e*	0.625(2)	0.4251(4)	0.7712(9)	0.009(9)	1
H2	4*e*	0.391(2)	0.5812(7)	0.5065(5)	0.009(9)	1
H3	4*e*	0.534(2)	0.7905(5)	0.6235(8)	0.009(9)	1
H4	4*e*	0.888(1)	0.5840(7)	0.7265(9)	0.009(9)	1
H5	4*e*	0.110(1)	0.7200(6)	0.6340(9)	0.009(9)	1
H6	4*e*	0.342(2)	0.5557(7)	0.8970(8)	0.009(9)	1
H7	4*e*	0.845(1)	0.5696(7)	1.0000(5)	0.009(9)	1
H8	4*e*	0.468(2)	0.7734(6)	0.8946(8)	0.009(9)	1
H9	4*e*	0.110(1)	0.4810(7)	0.6235(9)	0.009(9)	1
H10	4*e*	0.936(1)	0.7718(7)	0.8900(9)	0.009(9)	1

Table S4. The refined $Na_2B_{10}H_{10}$ structural parameters at 300 K associated with the monoclinic space group $P2_1/c$ (No. 14) derived from XRPD data (R_p=0.0836, R_{wp}=0.0715, χ^2=0.332).

$Na_2B_{10}H_{10}$		**T = 300 K**			**XRPD**	**λ=0.7296(1) Å**
a = 6.7333(3) Å;	b= 13.1637(6) Å;	c = 11.9761(4) Å;	γ=120.591(2) °;	V = 913.76(7) Å^3;	Z = 4	
Atom	Site	x	y	z	U_{iso} (Å^2)	Occ.
Na1	4*e*	0.234(1)	0.3896(7)	0.4803(7)	0.035(1)	1
Na2	4*e*	0.205(1)	0.6344(7)	0.0060(8)	0.035(1)	1
B1	4*e*	0.562(1)	0.5134(4)	0.7615(7)	0.006(1)	1
B2	4*e*	0.436(1)	0.5962(5)	0.6173(6)	0.006(1)	1
B3	4*e*	0.540(1)	0.7179(5)	0.6892(7)	0.006(1)	1
B4	4*e*	0.735(1)	0.6101(6)	0.7454(7)	0.006(1)	1
B5	4*e*	0.284(1)	0.6671(5)	0.6823(7)	0.006(1)	1
B6	4*e*	0.409(1)	0.5851(5)	0.8264(7)	0.006(1)	1
B7	4*e*	0.712(1)	0.6028(5)	0.8920(6)	0.006(1)	1
B8	4*e*	0.518(1)	0.7102(5)	0.8378(7)	0.006(1)	1
B9	4*e*	0.274(1)	0.5395(5)	0.6724(7)	0.006(1)	1
B10	4*e*	0.774(1)	0.7131(5)	0.8411(7)	0.006(1)	1
H1	4*e*	0.626(2)	0.4271(4)	0.773(1)	0.011(9)	1
H2	4*e*	0.394(2)	0.5811(8)	0.5086(6)	0.011(9)	1
H3	4*e*	0.535(2)	0.7886(6)	0.6248(8)	0.011(9)	1
H4	4*e*	0.888(1)	0.5843(7)	0.729(1)	0.011(9)	1
H5	4*e*	0.112(1)	0.7185(7)	0.6330(9)	0.011(9)	1
H6	4*e*	0.342(2)	0.5567(7)	0.8954(8)	0.011(9)	1
H7	4*e*	0.843(1)	0.5707(7)	1.0001(6)	0.011(9)	1
H8	4*e*	0.467(2)	0.7721(6)	0.8927(9)	0.011(9)	1
H9	4*e*	0.114(1)	0.4818(7)	0.623(1)	0.011(9)	1
H10	4*e*	0.934(1)	0.7707(7)	0.891(1)	0.011(9)	1

Table S5. The refined $Na_2{}^{11}B_{10}D_{10}$ structural parameters at 410 K associated with the cubic space group *Fm*-3*m* (No. 225) derived from NPD data (R_{wp}=0.0491, R_p=0.0412, χ^2=1.403).

$Na_2{}^{11}B_{10}D_{10}$		**T = 410 K**			**NPD Cu(311)/λ=1.5398(2) Å**	
a = 9.8397(5) Å; *V* = 952.7(1) Å^3; Z = 4						
Atom	Site	*x*	*y*	*z*	U_{iso} (Å^2)	Occ.
B1	24*e*	0.1864(4)	0	0	0.119(6)	⅓
B2	96*k*	0.0634(4)	0.0625(4)	0.1279(7)	0.135(6)	⅓
D1	24*e*	0.3058(4)	0	0	0.35(1)	⅓
D2	96*k*	0.1103(4)	0.1105(4)	0.226(2)	0.35(1)	⅓
Na1	8*c*	¼	¼	¼	0.09	0.54(2)
Na2	4*b*	½	0	0	0.09	0.11(3)
Na3	24*d*	¼	¼	0	0.19	0.10(1)

Table S6. The refined $Na_2B_{10}H_{10}$ structural parameters at 410 K associated with the cubic space group *Fm*-3*m* (No. 225) derived from synchrotron XRPD data (R_p=0.136, R_{wp}=0.0924, χ^2=0.482).

$Na_2B_{10}H_{10}$		**T = 410 K**			**XRPD λ=0.7296(1) Å**	
a = 9.8517(6) Å; *V* = 956.16(9) Å^3; Z = 4						
Atom	Site	*x*	*y*	*z*	U_{iso} (Å^2)	Occ.
B1	24*e*	0.1852(6)	0	0	0.05	⅓
B2	96*k*	0.0632(3)	0.0632(3)	0.1284(4)	0.23	⅓
Na1	8*c*	¼	¼	¼	0.27	0.742(5)
Na2	4*b*	½	0	0	0.60	0.06(1)
Na3	24*d*	¼	¼	0	0.56	0.056(8)

N.B.: H atom positions were ignored during the refinement.

Table S7. List of phonon symmetries and the corresponding phonon energies [Symmetry, Energy (meV), Energy (cm^{-1})] at the gamma point from the DFT-optimized $Na_2{}^{11}B_{10}H_{10}$ structure at 0 K. All modes are nondegenerate. Modes below 200 cm^{-1} possess cation and whole-anion translational and/or librational character. (N.B.: Infrared-active modes = Au, Bu and Raman-active modes = Ag, Bg.)

Sym	E(mV)	E(cm^{-1})
Au	3.6	29.21
Ag	5.2	42.00
Ag	7.6	61.01
Bg	7.7	62.39
Au	8.2	66.13
Bu	8.5	68.59
Ag	8.7	69.91
Au	8.7	70.28
Bg	8.9	72.10
Au	9.8	78.67
Bu	9.8	79.14
Ag	10.5	85.03
Bg	10.6	85.46
Bu	10.7	86.25
Ag	10.8	87.12
Au	10.9	88.26
Bg	11.9	95.90
Bg	12.3	99.36
Ag	12.4	99.76
Au	12.6	101.59
Ag	13.2	106.63
Bu	13.3	107.24
Bg	14.4	115.98
Ag	14.5	117.15
Bu	14.7	118.83
Bu	15.4	123.95
Bg	16.0	129.30
Bu	16.3	131.65
Ag	16.4	132.04
Bg	16.4	132.51
Au	17.1	137.96
Bg	17.8	143.75
Bu	18.0	145.36
Au	18.3	147.34
Ag	18.6	150.21
Au	19.1	154.26
Bg	19.4	156.35
Bg	20.5	165.54
Bu	21.1	169.95
Ag	21.5	173.59
Au	21.8	175.92
Au	22.0	177.13
Ag	23.4	188.41
Bg	23.6	190.16
Bu	24.4	196.60
Bu	56.8	458.03
Au	57.3	461.92
Bg	57.6	464.88
Ag	57.7	465.48
Au	58.7	473.54
Ag	58.9	475.33
Bu	59.1	476.45
Bg	59.1	476.70
Au	59.1	476.99
Bg	59.4	478.91
Bu	59.4	478.99
Ag	59.5	480.25
Bu	65.9	531.88
Bu	66.0	532.42
Ag	66.0	532.45
Au	66.1	533.08
Bg	66.2	533.71
Au	66.3	534.75
Bg	66.6	536.97
Ag	66.7	537.82
Au	71.2	574.10
Bg	71.7	578.38
Bu	71.7	578.81
Ag	72.1	581.85
Au	72.5	584.99
Bu	72.9	588.22
Ag	72.9	588.51
Bg	73.3	591.09
Bu	75.6	609.77
Bg	75.6	610.20
Au	75.7	610.78
Ag	75.9	612.43
Bu	76.6	618.21
Bg	76.8	619.85
Au	76.9	620.08
Ag	76.9	620.73
Bu	77.0	621.46
Au	77.2	622.55
Bg	77.3	623.55
Ag	77.4	624.70
Bu	81.9	660.86
Au	82.2	662.84
Bg	82.3	663.83
Ag	82.3	663.92
Bu	82.4	664.46
Ag	82.5	665.74
Au	82.5	665.76
Bg	82.7	667.51
Au	82.8	668.15
Bu	83.0	669.57
Ag	83.4	672.61
Bg	83.5	673.24
Bu	84.1	678.65
Au	84.5	681.41
Ag	84.5	682.10
Bg	84.7	683.21
Au	86.6	698.98
Bg	86.8	700.62
Bu	86.9	700.94
Ag	87.0	701.73
Au	87.5	706.06
Bu	88.1	710.76
Bg	88.3	712.07
Ag	88.3	712.71
Au	90.5	729.71
Bu	90.5	729.78
Ag	91.1	734.98
Bg	91.1	735.09
Au	91.5	738.35
Bu	91.6	738.93
Ag	91.8	740.59
Bg	91.9	741.48
Bu	92.2	744.06
Au	92.3	744.56
Bg	92.7	747.85
Ag	92.8	748.83
Au	93.5	754.61
Bg	94.0	758.11
Bu	94.2	759.69
Ag	94.5	762.55
Bu	96.4	777.66
Bg	96.4	777.76
Ag	96.7	780.25
Au	96.9	781.52
Ag	97.5	786.66
Au	97.6	787.60
Bu	97.7	788.25
Bg	97.8	788.68
Au	98.9	797.85
Bg	99.1	799.13
Ag	99.1	799.24
Bu	99.1	799.66
Au	102.7	828.73
Bu	102.9	830.22
Bg	103.1	831.38
Ag	103.1	831.54
Bu	103.7	836.82
Bg	103.9	837.87
Ag	103.9	838.04
Au	104.0	838.81
Ag	106.3	857.21
Au	106.4	858.35
Bu	106.5	859.13
Bg	106.7	860.92
Bu	106.8	861.84
Au	106.9	862.47
Bg	107.8	869.88
Ag	107.8	869.90
Au	107.8	870.08
Bg	108.1	872.05
Bu	108.2	872.94
Ag	108.3	873.99
Bu	108.9	878.51
Au	109.0	879.61
Bg	109.2	880.70
Ag	109.2	880.78
Au	110.2	888.77
Bu	110.4	890.48
Au	110.7	892.84
Bu	110.7	892.96
Bg	110.7	893.11
Ag	110.7	893.34
Au	110.9	894.63
Bu	110.9	895.01
Bg	111.0	895.43
Ag	111.0	895.59
Bg	111.1	896.17
Ag	111.2	897.08
Bu	112.2	904.88
Au	112.4	906.76
Bg	112.4	906.80
Ag	112.6	908.02
Au	112.8	909.77
Bu	112.8	910.41
Ag	112.9	910.69
Bg	113.1	912.79
Au	114.3	921.81
Bu	114.4	922.60
Bg	114.4	922.87
Ag	114.5	923.47
Bu	114.8	926.26
Ag	114.8	926.44
Au	114.9	926.75
Bg	114.9	926.79
Bu	115.9	934.71
Au	115.9	935.32
Ag	116.0	935.43
Bg	116.0	935.45
Au	119.3	962.12
Ag	119.3	962.14
Bu	119.3	962.80
Bg	119.4	963.29
Au	119.8	966.36
Ag	119.8	966.48
Bg	119.8	966.70
Bu	119.9	967.41
Bu	120.2	969.64
Bg	120.3	970.44
Ag	120.4	971.55
Au	120.5	971.83
Bu	125.8	1014.93
Au	125.9	1016.06
Ag	126.0	1016.25
Bg	126.1	1016.98
Au	126.4	1019.93
Bu	126.5	1020.21
Ag	126.5	1020.51
Bg	126.6	1021.72
Ag	127.0	1024.68
Au	127.0	1024.71
Bu	127.1	1025.02
Bg	127.2	1025.84
Bu	133.5	1076.79
Au	133.5	1077.00
Ag	133.6	1077.75
Bg	133.6	1077.77
Bu	306.6	2473.28
Au	306.6	2473.49
Bg	307.9	2483.87
Ag	307.9	2484.27
Au	308.4	2487.72
Bu	308.4	2487.77
Ag	308.5	2488.85
Bg	308.7	2490.15
Bu	309.5	2496.94
Au	310.0	2500.58
Ag	310.5	2505.28
Bg	310.6	2505.98
Bu	313.3	2527.86
Ag	313.4	2528.49
Au	313.8	2531.92
Bg	314.0	2533.59
Bu	314.1	2534.20
Au	314.2	2534.77
Ag	314.2	2534.92
Bg	314.3	2535.92
Au	315.0	2541.01
Bu	315.0	2541.60
Bg	315.1	2542.33
Ag	315.1	2542.47
Bu	315.3	2543.55
Au	315.5	2545.20
Ag	315.6	2545.99
Au	315.7	2547.02
Bg	315.8	2547.49
Bu	316.3	2551.47
Ag	316.4	2552.33
Bu	316.6	2553.97
Au	316.7	2555.12
Bg	316.8	2556.14
Ag	317.3	2559.46
Bg	317.3	2559.65
Au	318.7	2571.40
Bu	318.8	2571.95
Bg	319.2	2575.01
Ag	319.3	2575.59

Table S8. List of phonon symmetries and the corresponding phonon energies [Symmetry, Energy (meV), Energy (cm^{-1})] at the gamma point from the DFT-optimized $Na_2{}^{11}B_{10}D_{10}$ structure at 0 K. All modes are nondegenerate. Modes below 200 cm^{-1} possess cation and whole-anion translational and/or librational character. (N.B.: Infrared-active modes = Au, Bu and Raman-active modes = Ag, Bg.)

Sym	E(mV)	E(cm^{-1})
Au	3.5	28.25
Ag	5.1	40.76
Ag	7.2	58.49
Bg	7.5	60.56
Au	7.9	63.86
Bu	8.1	64.97
Au	8.3	66.67
Ag	8.4	67.65
Bg	8.6	69.18
Au	9.2	74.50
Bu	9.6	77.07
Ag	10.1	81.13
Bg	10.3	82.75
Bu	10.3	83.36
Ag	10.4	83.63
Au	10.9	87.76
Bg	11.5	92.86
Au	12.0	96.67
Bg	12.2	98.38
Ag	12.2	98.67
Bu	12.7	102.69
Ag	12.9	104.47
Bg	14.0	113.04
Ag	14.4	116.23
Bu	14.6	117.79
Bu	14.9	120.52
Bu	15.5	124.95
Bg	15.5	125.35
Ag	15.8	127.12
Bg	15.8	127.56
Au	16.8	135.80
Bu	17.3	139.36
Bg	17.5	141.11
Au	17.5	141.52
Ag	17.8	143.52
Au	18.4	148.76
Bg	18.5	149.46
Bg	19.7	158.77
Ag	20.5	165.16
Bu	20.7	166.84
Au	21.1	169.83
Au	21.8	175.61
Bg	22.6	182.05
Ag	22.7	183.42
Bu	23.7	191.06
Bu	51.8	417.53
Au	52.1	420.72
Bg	52.7	425.46
Ag	52.8	425.61
Au	53.2	428.82
Bu	53.4	430.70
Bg	53.5	431.24
Ag	53.5	431.26
Au	53.6	432.76
Ag	53.9	435.19
Bu	54.0	435.31
Bg	54.0	435.62
Au	58.1	468.74
Bu	58.2	469.36
Bu	58.4	471.10
Ag	58.5	471.79
Au	58.6	472.84
Bg	58.6	473.10
Bg	59.1	476.78
Ag	59.4	479.41
Au	60.1	485.08
Bg	60.2	486.03
Bu	60.4	487.12
Au	60.5	488.31
Bu	60.7	490.06
Ag	60.8	490.82
Ag	61.0	492.52
Bg	61.5	496.47
Bu	63.9	515.90
Au	64.3	518.81
Au	64.8	522.66
Ag	64.8	523.17
Bg	65.3	526.64
Bu	65.4	527.33
Bg	65.5	528.36
Ag	65.5	528.45
Au	66.4	535.28
Bu	66.4	535.67
Au	66.4	535.73
Ag	66.6	537.45
Bg	66.6	537.60
Bu	66.8	538.64
Bg	66.9	539.92
Bu	67.2	541.81
Ag	67.2	542.37
Bg	67.3	543.29
Ag	67.5	544.53
Au	67.5	544.71
Bu	68.8	554.74
Bg	68.8	554.83
Ag	68.8	555.06
Au	68.9	555.50
Au	69.8	562.81
Bu	69.8	563.28
Ag	69.8	563.44
Bg	69.9	563.68
Au	71.1	573.56
Ag	71.1	573.68
Bg	71.1	573.75
Bu	71.2	574.52
Bu	72.8	587.28
Au	72.8	587.28
Bg	72.9	587.77
Ag	72.9	588.06
Bu	74.9	604.50
Bg	75.0	604.99
Ag	75.1	605.54
Bu	75.1	605.81
Au	75.1	606.04
Au	75.1	606.17
Bg	75.2	606.67
Ag	75.2	606.82
Au	76.2	614.76
Bu	76.4	616.75
Bg	76.5	616.87
Ag	76.8	619.59
Bu	78.1	629.96
Bg	78.1	630.07
Au	78.2	631.17
Ag	78.3	631.37
Bg	82.1	662.22
Ag	82.1	662.25
Bu	82.1	662.26
Au	82.1	662.31
Bu	82.6	666.46
Ag	82.7	667.28
Au	82.7	667.36
Bg	82.7	667.55
Bu	84.5	681.50
Au	84.6	682.76
Bg	84.6	682.88
Ag	84.7	683.03
Au	85.3	687.81
Bu	85.3	688.39
Ag	85.4	688.71
Bg	85.4	688.83
Au	86.6	698.82
Bu	86.7	699.25
Bg	86.8	699.90
Ag	86.8	700.44
Au	87.0	702.04
Bu	87.1	703.07
Bg	87.3	704.55
Ag	87.3	704.59
Au	87.5	705.67
Bu	87.6	706.34
Ag	87.6	706.81
Bg	87.7	707.76
Au	88.0	709.55
Ag	88.0	710.26
Bg	88.1	710.69
Bu	88.1	710.81
Bu	88.6	715.14
Ag	88.7	715.35
Au	88.7	715.54
Bg	88.7	715.63
Bu	89.8	724.80
Ag	89.8	724.82
Au	89.9	724.98
Bg	89.9	725.53
Bu	90.7	731.88
Ag	90.7	732.12
Au	90.8	732.39
Bg	90.8	732.77
Ag	91.7	740.03
Bg	91.7	740.11
Bu	91.8	740.40
Au	91.8	740.44
Bg	91.9	741.63
Bu	91.9	741.63
Au	91.9	741.65
Ag	92.0	741.90
Bg	94.2	759.68
Au	94.2	759.83
Bu	94.2	760.13
Ag	94.6	762.81
Bu	96.4	777.88
Au	96.4	778.00
Bg	96.5	778.36
Ag	96.5	778.42
Bu	97.3	785.31
Au	97.4	785.47
Ag	97.4	785.82
Bg	97.4	786.12
Au	106.6	859.74
Bu	106.6	859.91
Ag	106.6	860.31
Bg	106.7	860.61
Bu	106.7	860.66
Au	106.7	860.71
Ag	106.7	861.09
Bg	106.8	861.28
Au	111.1	896.05
Bg	111.1	896.18
Bu	111.1	896.38
Ag	111.1	896.51
Bu	111.3	897.87
Bg	111.3	897.91
Au	111.3	898.10
Ag	111.3	898.26
Au	112.6	908.76
Ag	112.7	909.60
Bu	112.8	909.97
Bg	112.8	910.27
Bu	113.1	912.29
Au	113.2	913.09
Ag	113.2	913.23
Bg	113.3	914.09
Bu	119.8	966.44
Bg	119.9	967.21
Au	119.9	967.44
Ag	120.0	967.71
Bu	121.9	983.61
Au	121.9	983.71
Ag	122.0	984.40
Bg	122.1	984.87
Bu	226.8	1829.78
Au	226.8	1829.99
Bg	227.7	1836.57
Ag	227.7	1836.91
Bu	228.0	1839.15
Au	228.0	1839.28
Ag	228.2	1840.84
Bg	228.2	1840.97
Bu	229.1	1848.58
Au	229.4	1850.99
Ag	229.7	1853.36
Bg	229.8	1853.64
Bu	231.5	1867.87
Ag	231.6	1868.69
Au	232.0	1872.06
Bg	232.1	1872.10
Au	232.2	1873.59
Ag	232.4	1874.54
Bu	232.4	1874.87
Bg	232.5	1875.53
Bu	233.0	1879.49
Bg	233.0	1879.66
Ag	233.1	1880.49
Au	233.1	1880.51
Bu	233.5	1883.77
Ag	233.6	1884.29
Bg	233.7	1885.36
Au	233.8	1885.81
Bu	233.9	1887.35
Au	234.0	1887.90
Bg	234.9	1894.67
Ag	234.9	1894.81
Au	235.1	1896.44
Ag	235.3	1898.52
Bu	235.4	1899.26
Bg	235.8	1902.18
Au	236.6	1908.87
Bu	236.6	1909.06
Bg	236.7	1909.80
Ag	236.9	1911.04