Supporting Materials

Dynamic Quantum Crystallography:

Lattice dynamical models refined against diffraction data

II. Applications to L-alanine, naphthalene and xylitol

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Table S0. Crystallographic details of the chosen systems and datasets.

Compound/temperature	**L-alanine 23K**	**Naphthalene 100K**	**Naphthalene 205K**	**Xylitol 122K**
Unit cell:	a=5.923Å b=12.260Å c=5.794Å	a=7.8248Å b=5.9349Å c=8.0997Å β=114.44°	a=7.9410Å b=5.9528Å c=8.1656Å β=115.1°	a=8.262 Å b=8.900 Å c=8.923 Å
$(\sin\theta/\lambda)_{max}$	1.08 $Å^{-1}$	1.15 $Å^{-1}$	1.13 $Å^{-1}$	1.1 $Å^{-1}$
R_{int}	Not given	0.0305	0.0321	0.017
Unique reflections	2533	4368	4250	7320
***R* after multipolar refinement**	0.0203	0.0177	0.0373	0.015
***wR* after multipolar refinement**	0.0159	0.0261	0.0364	0.024
Reference	The Journal of Physical Chemistry, Vol. 92, No. 4, 1988	J. Phys. Chem. A, Vol. 108, No. 6, 2004, 1059	J. Phys. Chem. A, Vol. 108, No. 6, 2004,1059	Acta Cryst. (2004). A60, 550±561

Table S1. Refinement statistics and indicators for all IAM (first left column) and normal mode models for tested datasets. *NrRefUij* stands for number of refined *Uij* parameters in IAM refinement, for *NoMoRe* it equals number of refined frequencies. In case of all statistics after *NoMoRe* refinements first value is a results of a refinement, for which frequencies from 6-31G(d,p) were used, whereas second, in italic were TZP.

Compound						
IAM (*SHELX*)			***NoMoRe***			
	Refined frequencies	**Maximum level of convergence (gradient norm)**	**wR2**	$\overline{S}_{nH}$	$\overline{S}_{H}$	**MFSF**
L-alanine 23K X-ray data						
wR2=0.082	**1 – 6**	$6.5*10^{-3}$/*$4*10^{-3}$*	0.0896/*0.0895*	0.09/*0.16*	0.31,0.54 /*0.27,0.49*	0.915/*0.905*
NrRefUij=36	**1 – 11**	$6.5*10^{-3}$/*$4.5*10^{-3}$*	0.0891/*0.0892*	0.10/*0.14*	0.33, 0.57/*0.27,0.45*	0.879/*0.855*
S_{TLS}=0.13	**1 - 16**	$5*10^{-3}$/*$8*10^{-3}$*	0.0895/*0.0892*	0.13/*0.14*	0.35,0.59/*0.30,0.54*	0.816/*0.891*
Naphthalene 100K X-ray data						
wR2=0.076	**1 – 11**	$1.2*10^{-2}$/*$1.7*10^{-2}$*	0.0824/*0.0843*	0.06/0.07	0.38/*0.43*	0.879/0.982
NrRefUij=30	**1 – 16**	$1.2*10^{-2}$/*$1.0*10^{-2}$*	0.0808/*0.0835*	0.06/0.06	0.42/*0.44*	1.067/1.01
Naphthalene 205K X-ray data						
wR2=0.091	**1 – 11**	$1.7*10^{-2}$/*$3*10^{-2}$*	0.1028/*0.1003*	0.05/*0.04*	0.31/*0.29*	0.977/*1.152*
NrRefUij=30	**1 – 16**	$1.7*10^{-2}$/*$1*10^{-2}$*	0.1027/*0.1018*	0.05/*0.04*	0.30/*0.22*	0.978/*0.825*
Naphthalene 80K neutron data						
wR2=0.113	**1 – 11**	$1.2*10^{-2}$/$5*10^{-3}$	0.1209/*0.1251*	0.32/*1.02*		0.665/0.825
NrRefUij=54	**1 – 16**	$1.2*10^{-2}$/$5*10^{-3}$	0.1227/*0.1245*	0.27/*0.98*		0.819/0.859
	1 – 21	$1.2*10^{-2}$/$5*10^{-3}$	0.1222/*0.1251*	0.25/*1.02*		0.944/1.001
Xylitol 122K X-ray data						
wR2=0.055	**1 – 11**	$1.2*10^{-2}$/*$2*10^{-2}$*	0.0691/*0.0712*	0.13/*0.16*	0.33/*0.29*	0.894/*0.881*
NrRefUij=60	**1 – 21**	$1.2*10^{-2}$/*$1*10^{-2}$*	0.0730/*0.7038*	0.16/*0.14*	0.36/*0.29*	0.762/*0.828*
S_{TLS}=0.29	**1 – 31**	$1.6*10^{-2}$/*$1*10^{-2}$*	0.0717/*0.0697*	0.16/*0.14*	0.33/*0.28*	0.756/*0.728*
Xylitol 122K neutron data						
wR2=0.053	**1 – 11**	$4*10^{-3}$/*$5*10^{-3}$*	0.0610/*0.0594*	0.27/*0.25*		0.833/*0.839*
NrRefUij=132	**1 – 21**	$3.5*10^{-3}$/*$8*10^{-3}$*	0.0612/*0.0560*	0.31/*0.27*		0.808/*0.781*
S_{TLS}=0.40	**1 – 31**	$6*10^{-3}$/*$8*10^{-3}$*	0.0611/*0.0592*	0.27/*0.22*		0.850/*0.810*

Table S2. Mean ADPs (first line) and Standard uncertainties (second line) on ADPs estimated via a Monte Carlo approach for xylitol. NoMoRe refinement using 22 parameters (including MFSF) against the 122 K X-ray dataset. The lattice-dynamical model is based on B3LYP/6-31G(d,p) calculations.

O1	0.01037	0.01558	0.01027	0.00092	-0.00124	0.00084
	0.00009	0.00021	0.00008	0.00004	0.00006	0.00007
O2	0.01017	0.01436	0.01102	-0.00060	-0.00024	-0.00297
	0.00011	0.00018	0.00008	0.00006	0.00005	0.00008
O3	0.01201	0.01190	0.00904	-0.00008	-0.00071	0.00033
	0.00011	0.00009	0.00012	0.00010	0.00008	0.00004
O4	0.01261	0.01091	0.01139	-0.00115	-0.00156	0.00142
	0.00012	0.00010	0.00012	0.00005	0.00006	0.00004
O5	0.00964	0.01573	0.01235	-0.00142	0.00110	-0.00261
	0.00012	0.00034	0.00009	0.00006	0.00004	0.00004
C1	0.00969	0.01748	0.00884	0.00070	-0.00005	0.00112
	0.00010	0.00025	0.00011	0.00009	0.00003	0.00004
C2	0.00791	0.01142	0.00836	-0.00056	-0.00085	-0.00042
	0.00013	0.00009	0.00011	0.00004	0.00009	0.00002
C3	0.00769	0.01093	0.00840	-0.00109	-0.00044	-0.00022
	0.00017	0.00010	0.00013	0.00008	0.00006	0.00006
C4	0.00776	0.01152	0.00836	-0.00029	-0.00034	-0.00042
	0.00017	0.00010	0.00010	0.00003	0.00003	0.00004
C5	0.00809	0.01156	0.01366	-0.00092	-0.00002	0.00005
	0.00011	0.00030	0.00021	0.00006	0.00004	0.00002
H1A	0.03913	0.03511	0.02359	-0.01090	-0.00524	0.01474
	0.00071	0.00068	0.00014	0.00027	0.00018	0.00058
H1B	0.01882	0.04997	0.03096	0.01722	0.00041	-0.00791
	0.00009	0.00120	0.00051	0.00077	0.00010	0.00025
H2	0.02255	0.01934	0.02290	-0.00570	-0.00040	0.00146
	0.00016	0.00009	0.00014	0.00007	0.00017	0.00009
H3	0.02292	0.02111	0.01977	-0.00634	0.00088	0.00147
	0.00011	0.00012	0.00009	0.00018	0.00004	0.00008
H4	0.02335	0.02862	0.01439	0.00329	-0.00063	-0.00191
	0.00012	0.00025	0.00010	0.00007	0.00002	0.00006
H5B	0.02490	0.03351	0.02520	0.01151	-0.00327	-0.00086
	0.00013	0.00051	0.00055	0.00068	0.00008	0.00006
H5A	0.02273	0.02349	0.04175	-0.01395	0.00234	0.00060
	0.00015	0.00018	0.00130	0.00051	0.00007	0.00009
H11	0.01810	0.02828	0.02157	0.00269	0.00286	-0.00102
	0.00010	0.00010	0.00013	0.00008	0.00006	0.00006
H12	0.02621	0.02834	0.01783	-0.00122	0.00573	-0.00527
	0.00012	0.00015	0.00009	0.00015	0.00004	0.00003
H13	0.02624	0.01725	0.02097	0.00139	-0.00072	-0.00117
	0.00010	0.00010	0.00010	0.00006	0.00005	0.00004
H14	0.02704	0.01934	0.02531	0.00303	-0.00216	0.00632
	0.00008	0.00010	0.00030	0.00008	0.00018	0.00003
H15	0.02482	0.02477	0.02056	-0.00439	-0.00089	-0.00648
	0.00015	0.00035	0.00015	0.00007	0.00009	0.00003

Table S3. Mean ADPs (first line) and Standard uncertainties (second line) on ADPs estimated via a Monte Carlo approach for naphthalene. NoMoRe refinement using 22 parameters (including MFSF) against the 100 K X-ray dataset. The lattice-dynamical model is based on B3LYP/6-31G(d,p) calculations.

C1	0.01216	0.01839	0.01509	0.00027	0.00310	-0.00005
	0.00006	0.00009	0.00010	0.00004	0.00006	0.00002
C2	0.01275	0.01315	0.01480	0.00074	0.00517	0.00244
	0.00006	0.00007	0.00010	0.00003	0.00005	0.00002
C3	0.01204	0.00960	0.01067	-0.00040	0.00505	0.00059
	0.00007	0.00007	0.00007	0.00001	0.00005	0.00001
C4	0.01624	0.01093	0.01255	-0.00205	0.00641	-0.00109
	0.00007	0.00007	0.00007	0.00004	0.00006	0.00003
C5	0.01553	0.01627	0.01313	-0.00250	0.00433	-0.00349
	0.00007	0.00008	0.00007	0.00005	0.00006	0.00005
H1	0.01946	0.04002	0.03408	0.00133	0.00018	0.00391
	0.00007	0.00014	0.00019	0.00009	0.00009	0.00004
H2	0.02999	0.02316	0.03828	-0.00143	0.01200	0.00954
	0.00007	0.00008	0.00019	0.00008	0.00008	0.00005
H4	0.03622	0.01893	0.03186	-0.00568	0.01450	0.00304
	0.00011	0.00008	0.00012	0.00007	0.00010	0.00006
H5	0.03237	0.03247	0.02677	-0.01007	0.00440	-0.00926
	0.00010	0.00012	0.00012	0.00011	0.00009	0.00010

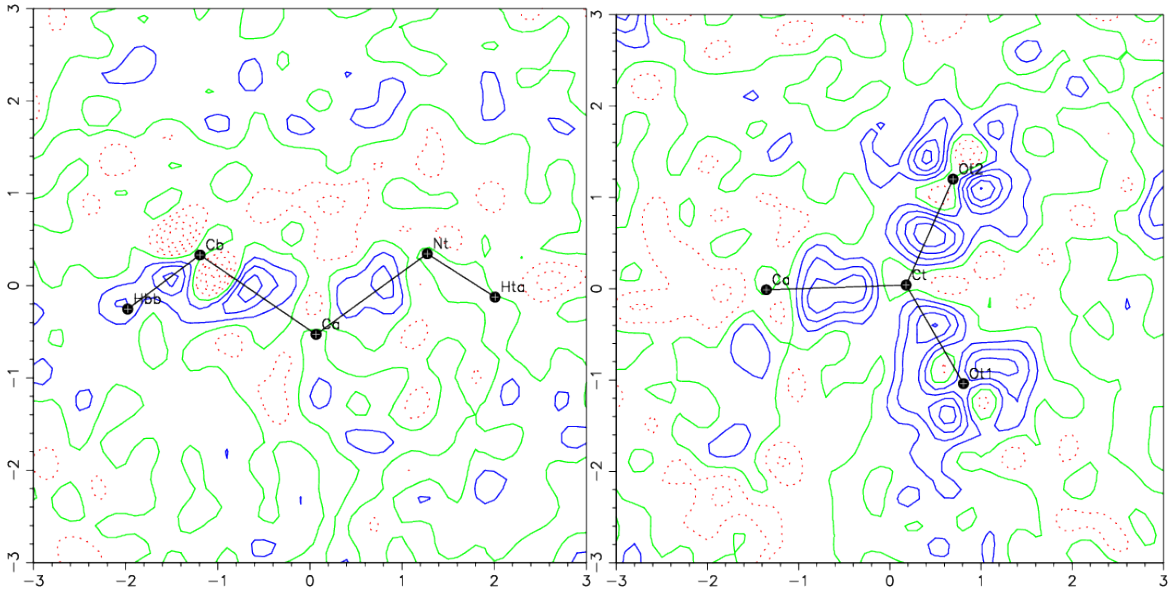

Figure 1S. Residual density for L-alanine. Left: the plane is defined by NT, CA and CB. Right: The plane is defined by OT1, CA and OT2. Contour level is 0.1e/Å^3. 15 frequencies were refined. B3LYP/6-31G(d,p) calculations.

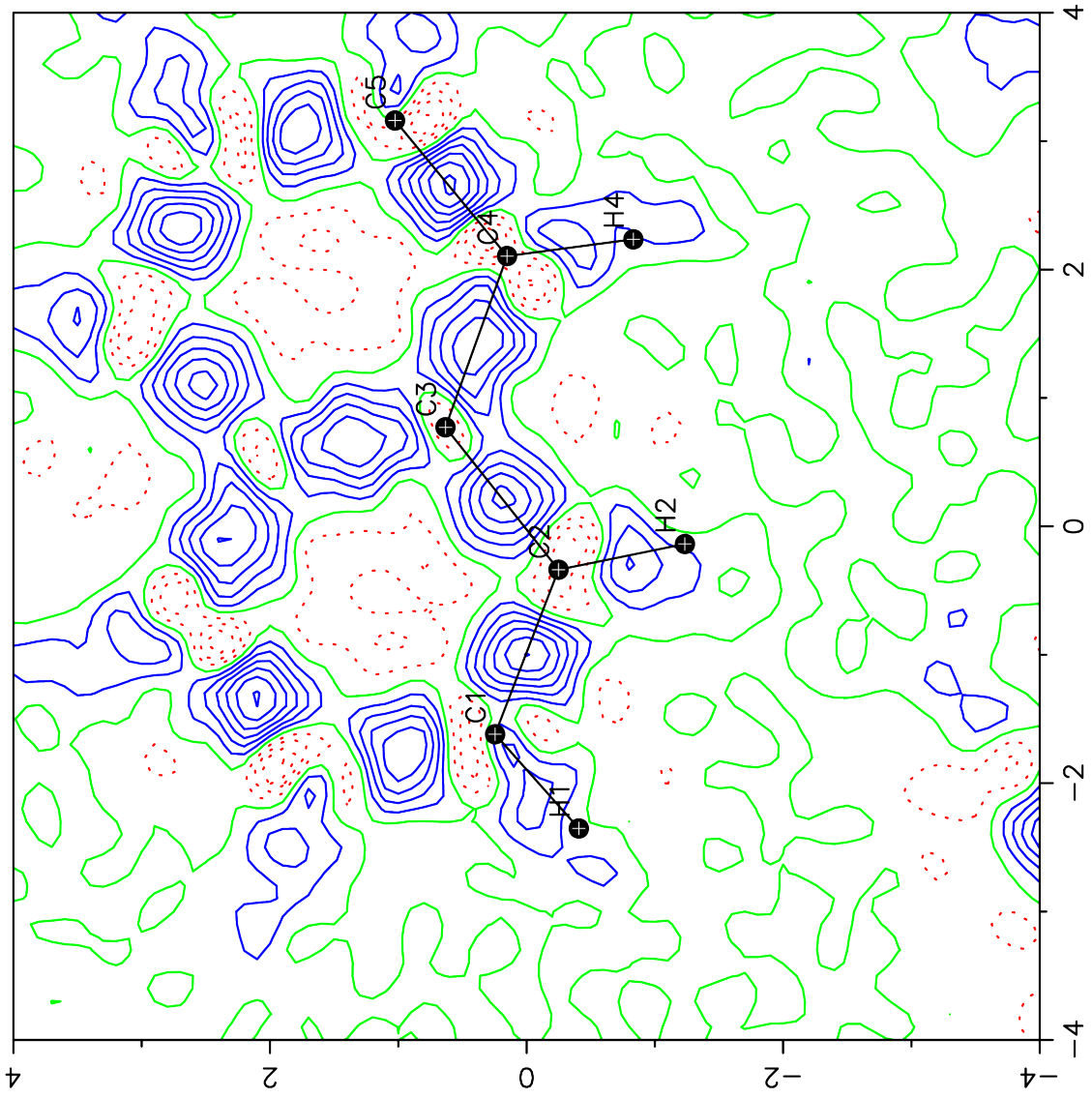

Figure 2S . Residual density for naphthalene. The plane is defined by C1, C2 and C5.Contour level is 0.1e/Å^3. 15 frequencies were refined. B3LYP/6-31G(d,p) calculations.

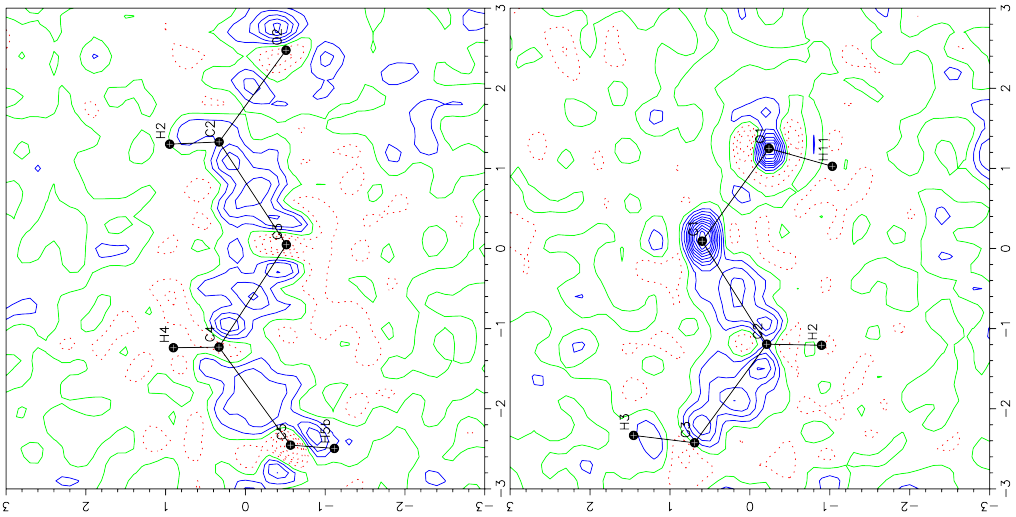

Figure 3S. Residual density for xylitol. The first plane is defined by C2, C3 and C4, and the second by O1, C1, C2. Contour level is 0.1e/Å^3. 21 frequencies were refined. B3LYP/6-31G(d,p) calculations.

Table S4. Final parameter values, estimated standard uncertainties and covariance matrix of selected refinements. These numbers are based on a Monte-Carlo approach to variance and co-variance estimation.

Napthalene 100 K X-ray data. 21 Parameters: MFSF and 20 frequency scaling factors. Based on B3LYP/TZP calculations. Correlation coefficients larger than 0.8 are colored yellow, and above 0.9 colored red.

Covariance matrix

```
[[ 1.    -0.1   -0.12  -0.13   0.06   0.15   0.03   0.26   0.38  -0.32   0.22  -0.12   0.18   0.47   0.5    0.04  -0.08   0.26   0.25   0.07   0.04   0.49]
 [-0.1    1.     0.15   0.2    0.03  -0.09  -0.17  -0.71  -0.05  -0.01   0.05  -0.06  -0.07  -0.15  -0.15  -0.02  -0.15  -0.15  -0.14  -0.06  -0.08  -0.06]
 [-0.12   0.15   1.     0.73   0.07  -0.81  -0.02  -0.07  -0.02   0.04  -0.34  -0.34  -0.27  -0.17  -0.17  -0.19  -0.22  -0.06  -0.09  -0.23  -0.18  -0.09]
 [-0.13   0.2    0.73   1.    -0.08  -0.73  -0.04  -0.09  -0.17   0.12  -0.49  -0.22  -0.28  -0.2   -0.2   -0.22  -0.23  -0.05  -0.08  -0.2   -0.16  -0.05]
 [ 0.06   0.03  -0.08  -0.08   1.    -0.01  -0.84   0.1    0.16   0.01  -0.04  -0.16   0.1    0.14   0.06   0.02  -0.06  -0.1   -0.18  -0.07  -0.07  -0.01]
 [ 0.15  -0.09  -0.81  -0.73  -0.01   1.     0.01   0.1    0.08  -0.04   0.3    0.18   0.27   0.13   0.16   0.23   0.24   0.05   0.06   0.15   0.13   0.11]
 [ 0.03  -0.17  -0.02  -0.04  -0.84   0.01   1.    -0.01   0.06  -0.18   0.17   0.04  -0.08  -0.    0.04  -0.09   0.04   0.09   0.17   0.04   0.04   0.  ]
 [ 0.26  -0.71  -0.07  -0.09   0.1    0.1   -0.01   1.     0.26  -0.2    0.05  -0.08   0.04   0.28   0.2    0.08  -0.05   0.06   0.04  -0.07  -0.06   0.1 ]
 [ 0.38  -0.05  -0.02  -0.17   0.16   0.08   0.06   0.26   1.    -0.85   0.43  -0.3    0.35   0.3   -0.18  -0.21  -0.04  -0.02  -0.07  -0.1    0.08]
 [-0.32  -0.01   0.04   0.12   0.01  -0.04  -0.18  -0.2   -0.85   1.    -0.43   0.23  -0.02  -0.35   0.17   0.15   0.05   0.01   0.06   0.06  -0.09]
 [ 0.22   0.05  -0.34  -0.49  -0.04   0.3    0.17   0.05  -0.43   1.     1.    -0.1    0.07   0.34   0.21   0.11  -0.03  -0.02   0.15   0.06   0.06]
 [-0.12  -0.06  -0.34  -0.22  -0.16   0.18   0.04  -0.08  -0.3    0.23  -0.1    1.    -0.02  -0.16  -0.05  -0.04   0.01   0.07   0.12  -0.02  -0.03  -0.01]
 [ 0.18  -0.07  -0.27  -0.28   0.1    0.27  -0.08   0.04  -0.02   0.03   0.07  -0.02   1.     0.12   0.06   0.2    0.08  -0.03  -0.04   0.09   0.07   0.04]
 [ 0.47  -0.15  -0.17  -0.2    0.14   0.13  -0.     0.28   0.35  -0.33   0.34  -0.16   0.12   1.     0.57   0.05   0.13   0.16   0.13   0.23   0.21   0.09]
 [ 0.5   -0.15  -0.17  -0.2    0.06   0.16   0.04   0.2    0.3   -0.35   0.21  -0.05   0.06   0.57   1.     0.01   0.14   0.28   0.25  -0.01   0.02   0.29]
 [ 0.04  -0.02  -0.19  -0.22   0.02   0.23  -0.09   0.08  -0.18   0.17   0.11  -0.04   0.2    0.05   0.01   1.     0.45   0.04   0.05   0.4    0.34   0.11]
 [-0.08  -0.15  -0.22  -0.23  -0.06   0.24   0.04  -0.05  -0.21   0.15  -0.03   0.01   0.08   0.13   0.14   0.45   1.     0.45   1.    -0.01   0.42   0.47   0.03]
 [ 0.26  -0.15  -0.06  -0.05  -0.1    0.05   0.09   0.06  -0.04   0.05  -0.02   0.07  -0.03   0.16   0.28   0.04  -0.01   1.     0.93   0.08   0.09   0.23]
 [ 0.25  -0.14  -0.09  -0.08  -0.18   0.06   0.17   0.04  -0.07   0.06   0.15  -0.02   0.08   0.13   0.25   0.05   0.    -0.93   1.     0.11   0.1    0.2 ]
 [ 0.07  -0.06  -0.23  -0.2   -0.07   0.15   0.04  -0.07  -0.07   0.06  -0.02   0.09   0.23  -0.01   0.4    0.42   0.08   0.11   1.     1.    -0.94  -0.06]
 [ 0.04  -0.08  -0.18  -0.16  -0.07   0.13   0.04  -0.06  -0.1    0.06  -0.03   0.07   0.21   0.02   0.34   0.47   0.09   0.1    0.94   1.    -0.07]
 [ 0.49  -0.06  -0.09  -0.05  -0.01   0.11   0.     0.1    0.08  -0.09   0.06  -0.01   0.04   0.09   0.11   0.03   0.23   0.2   -0.06  -0.07   1.  ]
]]
```

#	mean	esd
0	1.02	0.02
1	0.80	0.01
2	0.78	0.01
3	0.71	0.01
4	1.26	0.02
5	0.98	0.02
6	0.91	0.01
7	0.95	0.02
8	1.04	0.04
9	0.74	0.02

#	mean	esd
10	1.00	0.04
11	0.79	0.03
12	0.92	0.03
13	1.04	0.03
14	1.02	0.03
15	0.96	0.03
16	0.97	0.02
17	1.02	0.03
18	1.00	0.02
19	0.99	0.02
20	0.99	0.02
21	1.00	0.02

Xylitol 122 K X-ray data. 21 Parameters: MFSF and 20 frequency scaling factors. Based on B3LYP/TZP calculations. Correlation coefficients larger than 0.8 are colored yellow, and above 0.9 colored red.

```
[[ 1.   -0.88 -0.9  -0.62 -0.01  0.87  0.69 -0.86 -0.86 -0.56 -0.87 -0.42  0.11 -0.04 -0.78 -0.71 -0.37  0.01  0.16 -0.16  0.07  0.63]
 [-0.88  1.    0.9   0.59  0.08 -0.82 -0.74  0.77  0.74  0.41  0.72  0.28 -0.26 -0.15  0.67  0.53  0.25 -0.07 -0.3   0.02 -0.23 -0.7 ]
 [-0.9   0.9   1.    0.64  0.15 -0.92 -0.78  0.85  0.8   0.41  0.77  0.26 -0.27 -0.16  0.69  0.56  0.23 -0.03 -0.35 -0.01 -0.25 -0.75]
 [-0.62  0.59  0.64  1.   -0.01 -0.61 -0.6   0.56  0.5   0.26  0.48  0.34 -0.22  0.07  0.57  0.44  0.1  -0.07 -0.06  0.08 -0.02 -0.39]
 [-0.01  0.08  0.15 -0.01  1.   -0.03 -0.03 -0.03 -0.02 -0.09 -0.07 -0.13  0.01 -0.14 -0.1  -0.22 -0.11  0.23 -0.4  -0.3  -0.06 -0.12]
 [ 0.87 -0.82 -0.92 -0.61 -0.03  1.    0.82 -0.83 -0.79 -0.46 -0.81 -0.29  0.22  0.15 -0.69 -0.67 -0.24  0.13  0.34 -0.12  0.18  0.72]
 [ 0.69 -0.74 -0.78 -0.6  -0.03  0.82  1.   -0.68 -0.68 -0.37 -0.62 -0.31  0.29  0.16 -0.53 -0.54 -0.13  0.25  0.28 -0.04  0.32  0.6 ]
 [-0.86  0.77  0.85  0.56 -0.03 -0.83 -0.68  1.    0.8   0.48  0.79  0.35 -0.17 -0.08  0.75  0.65  0.37 -0.02 -0.15  0.09 -0.16 -0.71]
 [-0.86  0.74  0.8   0.5  -0.02 -0.79 -0.68  0.8   1.    0.6   0.75  0.49 -0.13  0.    0.68  0.67  0.36 -0.03 -0.11  0.15 -0.17 -0.59]
 [-0.56  0.41  0.41  0.26 -0.09 -0.46 -0.37  0.48  0.6   1.    0.41  0.35  0.14  0.08  0.6   0.66  0.34 -0.09  0.2   0.41  0.05 -0.22]
 [-0.87  0.72  0.77  0.48 -0.07 -0.81 -0.62  0.79  0.75  0.41  1.    0.38 -0.01  0.09  0.68  0.7   0.4   0.03 -0.14  0.17 -0.   -0.57]
 [-0.42  0.28  0.26  0.34 -0.13 -0.29 -0.31  0.35  0.49  0.35  0.38  1.    0.16  0.32  0.39  0.44  0.33  0.01  0.19  0.17  0.17 -0.06]
 [ 0.11 -0.26 -0.27 -0.22  0.01  0.22  0.29 -0.17 -0.13  0.14 -0.01  0.16  1.    0.22 -0.    0.14  0.11  0.25  0.18  0.16  0.41  0.32]
 [-0.04 -0.15 -0.16  0.07 -0.14  0.15  0.16 -0.08  0.    0.08  0.09  0.32  0.22  1.    0.08  0.1   0.29  0.03  0.31  0.23  0.27  0.25]
 [-0.78  0.67  0.69  0.57 -0.1  -0.69 -0.53  0.75  0.68  0.6   0.68  0.39 -0.    0.08  1.    0.68  0.42 -0.12  0.12  0.22  0.04 -0.51]
 [-0.71  0.53  0.56  0.44 -0.22 -0.67 -0.54  0.65  0.67  0.66  0.7   0.44  0.14  0.1   0.68  1.    0.38 -0.13  0.14  0.4   0.17 -0.36]
 [-0.37  0.25  0.23  0.1  -0.11 -0.24 -0.13  0.37  0.36  0.34  0.4   0.33  0.11  0.29  0.42  0.38  1.    0.06  0.17  0.34  0.23 -0.13]
 [ 0.01 -0.07 -0.03 -0.07  0.23  0.13  0.25 -0.02 -0.03 -0.09  0.03  0.01  0.25  0.03 -0.12 -0.13  0.06  1.    0.06 -0.03  0.12  0.15]
 [ 0.16 -0.3  -0.35 -0.06 -0.4   0.34  0.28 -0.15 -0.11  0.2  -0.14  0.19  0.18  0.31  0.12  0.14  0.17  0.06  1.    0.34  0.35  0.39]
 [-0.16  0.02 -0.01  0.08 -0.3  -0.12 -0.04  0.09  0.15  0.41  0.17  0.17  0.16  0.23  0.22  0.4   0.34 -0.03  0.34  1.    0.22  0.16]
 [ 0.07 -0.23 -0.25 -0.02 -0.06  0.18  0.32 -0.16 -0.17  0.05 -0.    0.17  0.41  0.27  0.04  0.17  0.23  0.12  0.35  0.22  1.    0.34]
 [ 0.63 -0.7  -0.75 -0.39 -0.12  0.72  0.6  -0.71 -0.59 -0.22 -0.57 -0.06  0.32  0.25 -0.51 -0.36 -0.13  0.15  0.39  0.16  0.34  1.  ]]
```

#	mean	esd
0	0.787	0.038
1	0.768	0.029
2	0.680	0.027
3	1.025	0.019
4	1.020	0.019
5	0.760	0.037
6	0.933	0.035
7	0.970	0.035
8	0.927	0.036
9	0.938	0.020

#	mean	esd
10	0.893	0.045
11	0.917	0.019
12	0.975	0.020
13	0.963	0.017
14	0.920	0.026
15	0.934	0.028
16	0.956	0.019
17	0.969	0.017
18	0.984	0.021
19	0.973	0.015
20	0.977	0.018
21	0.989	0.025

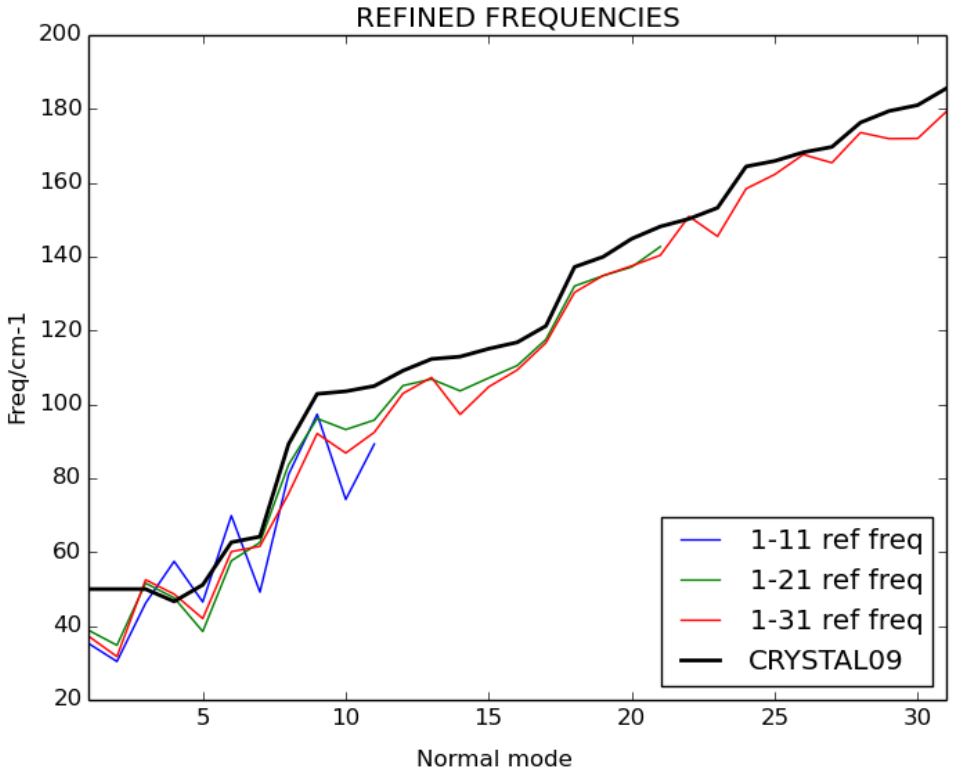

Figure S4. Frequencies for xylitol for particular normal modes obtained after *NoMoRe.* Refinement against X-ray 122 K data. Calculations at the B3LYP/6-31G(d,p) level.

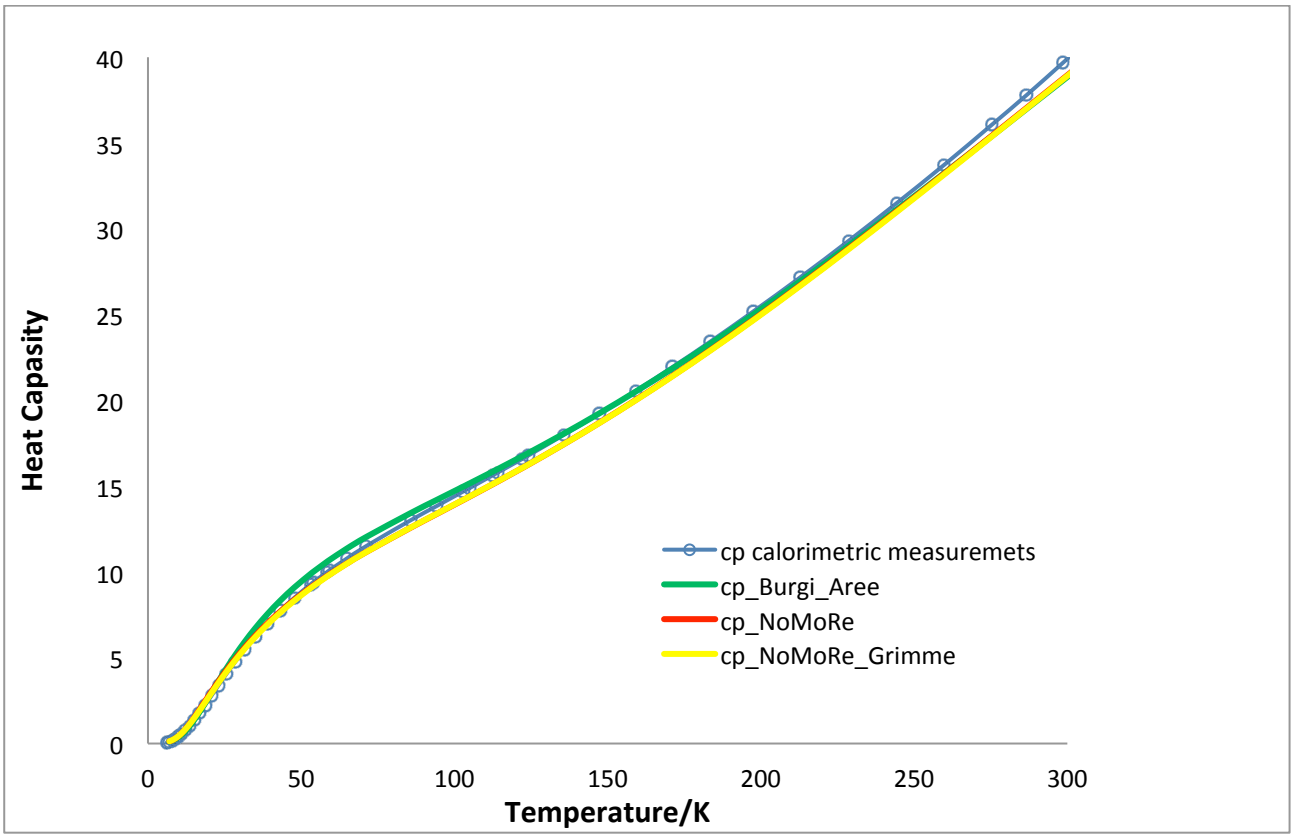

Figure S5. Heat capacity for naphthalene after *NoMoRe (red and yellow curves)* compared with calorimetric measurements. Red and yellow curves – comparison calculations with/without Grimme dispersion.