Supplementary Information: Using forces to accelerate first-principles anharmonic vibrational calculations

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1 Pseudopotentials

All density functional theory calculations were performed using CASTEP version 8.0, and its own "on-the-fly" ultrasoft pseudopotentials. The definition strings for the pseudopotentials were:

- H:

  `1|0.6|1|6|10|10(qc=8)`

- Li:

  `1|1.0|14|16|18|10U:20(qc=7)`

- Zr:

  `3|2.1|7|8|9|40U:50:41:42`

2 Equilibrium unit cell configurations

The unit cells for the structures used in this work, containing the atoms at their equilibrium positions, are given in the `.cif` files `H2.cif`, `cmca4.cif`, `cmca12.cif`, `c2c24.cif`, `Li.cif` and `Zr.cif`.

3 Harmonic mode displacement patterns

The displacement patterns corresponding to the mapping directions used in the mapping of 2-D subspaces of the BO surface of the *Cmca*-4 structure of solid hydrogen are given below. They correspond to the displacement patterns of harmonic modes with frequencies of 69.4, 74.0 and 114 meV, labelled as 4,

5 and 7 respectively. Each row shows the displacement of a H atom in the three Cartesian directions, in the same order as the atoms are listed in the file `cmca4.cif`.

- Direction 4:

```
x     y     z
0.00  0.75 -1.00
0.00  0.75  1.00
0.00 -0.75  1.00
0.00 -0.75 -1.00
```

- Direction 5:

```
x  y  z
0  1  0
0 -1  0
0  1  0
0 -1  0
```

- Direction 7:

```
 x  y  z
 1  0  0
-1  0  0
-1  0  0
 1  0  0
```