

TABLE S1

Force constants for the $[\text{ZnH}_4]^{2-}$ ion in D_{2d} symmetry.

| Value* | Description |
|--------|--|
| 1.130 | Zn-H stretch |
| -0.049 | Zn-H/Zn-H stretch interaction with 110° between bonds |
| -0.009 | Zn-H/Zn-H stretch interaction with 107° between bonds |
| 0.288 | H-Zn-H bend |
| -0.097 | Bend interaction where bond angle is 107° and 107° . |
| -0.015 | Bend interaction where bond angle is 110° and 110° . |
| -0.012 | Bend interaction where bond angle is 107° and 110° . |

*Diagonal force constants have units of $\text{mdyne}/\text{\AA}^{-1}$ and interaction force constants have units of $\text{mdyne}/\text{\AA}^{-2}$

All forcefield analysis used a general valence forcefield (GVFF) where the bondlengths and angles are taken from the crystal structures^{7,8} (and are also given in the Introduction). Internal coordinates are: bond stretch M-H and bond bending H-M-H. For inclusion of the librational modes, point masses were located around the complex and torsions defined about the M-H bonds. Symmetry coordinates were constructed from the internal coordinates using the in-house program SCORD which uses the conventional projection operator method (see F. A. Cotton, Chemical Applications of Group Theory, Wiley for details). The output from CLIMAX does not include uncertainties in the force constants, however, by varying the force constants manually, it is estimated that the uncertainty is *ca* 5%.

TABLE S2

Force constants for the $[\text{NiH}_4]^{2-}$ ion in C_1 symmetry.

| Value* | Description |
|--------|---|
| 1.670 | Ni-H stretch |
| 0.035 | Ni-H/Ni-H stretch interaction |
| 0.235 | H-Ni-H bend |
| -0.081 | Bend interaction where one bond angle includes the long |
| | (1.5718 \AA) bond. |
| -0.037 | Bend interaction that does not include the long |
| | (1.5718 \AA) bond. |

*Diagonal force constants have units of $\text{mdyne}/\text{\AA}^{-1}$ and interaction force constants have units of $\text{mdyne}/\text{\AA}^{-2}$