**Supporting Information for:**

**Structure-dependent vibrational dynamics in Mg(BH4)2 polymorphs probed with neutron vibrational spectroscopy and first-principles calculations**

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**How to view phonon animations using the V\_Sim software\***

The **alphaMg(BH4)2.xyz**, **gammaMg(BH4)2.xyz,** and **deltaMg(BH4)2.xyz** files contain the information needed to view the animated (gamma-point) phonon normal modes from the DFT-optimized 0 K α‑Mg(BH4)2, β-Mg(BH4)2, and δ-Mg(BH4)2 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://inac.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 **alphaMg(BH4)2.xyz, gammaMg(BH4)2.xyz,** or **deltaMg(BH4)2.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 mode energies are indicated in wavenumbers cm-1 (1 meV ≈ 8.066 cm-1).

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