Supporting information

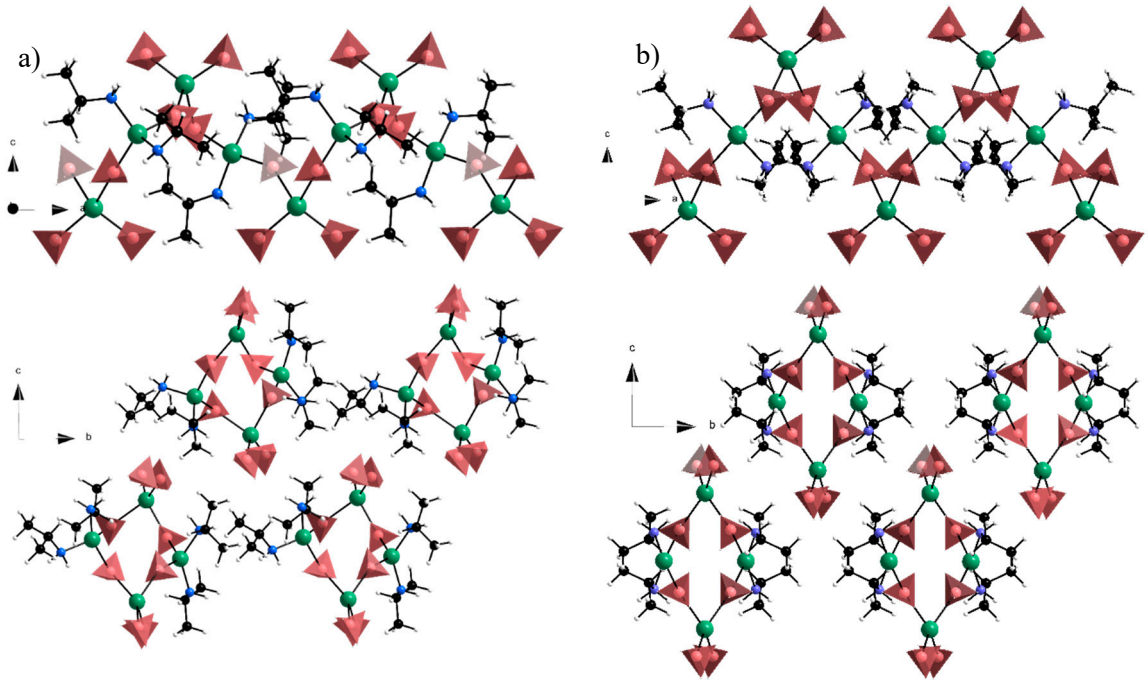

Figure S1. a) Most stable structure of $Mg(BH_4)_2{\cdot}(CH_3)_2CHNH_2$ found from DFT optimization. b) Metastable DFT-optimized structure of $Mg(BH_4)_2{\cdot}(CH_3)_2CHNH_2$ that resembles the experimental structure.

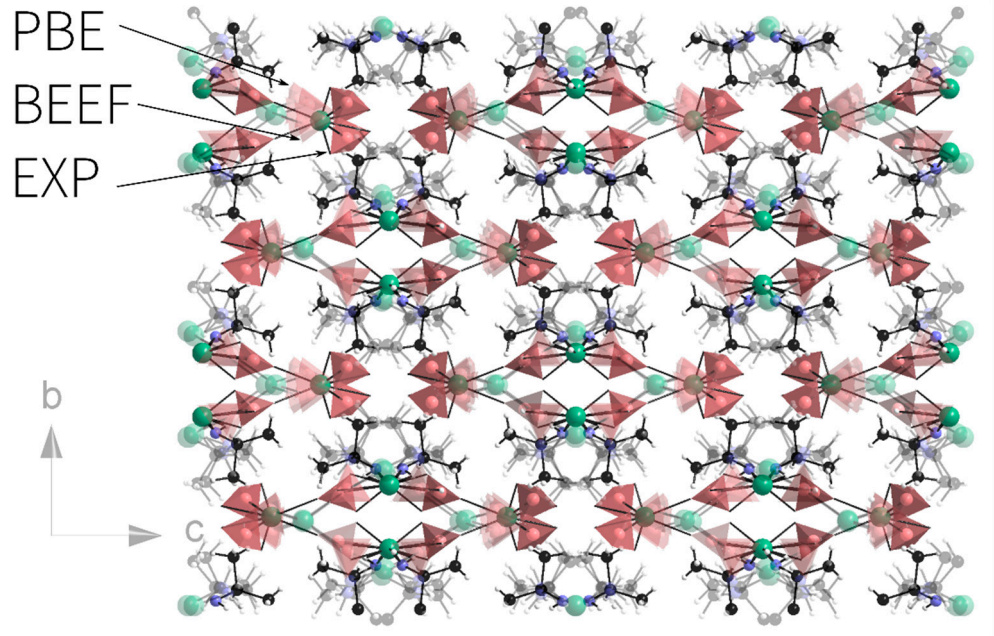

Figure S2. Comparison between the metastable structure found from DFT (PBE as well as BEEF-vdW functional) and the experimental structure. While the bonding schemes remain the same, Mg-Mg distances are longer for the experimental structure as terminal BH_4^- is closer to the isopropyl groups, suggesting that a B-$H^{\delta-}$… $^{0\delta}$H-C interaction exists. Accounting for vdW interaction with the BEEF-vdW functional increases the fit between DFT and experimental structure slightly.

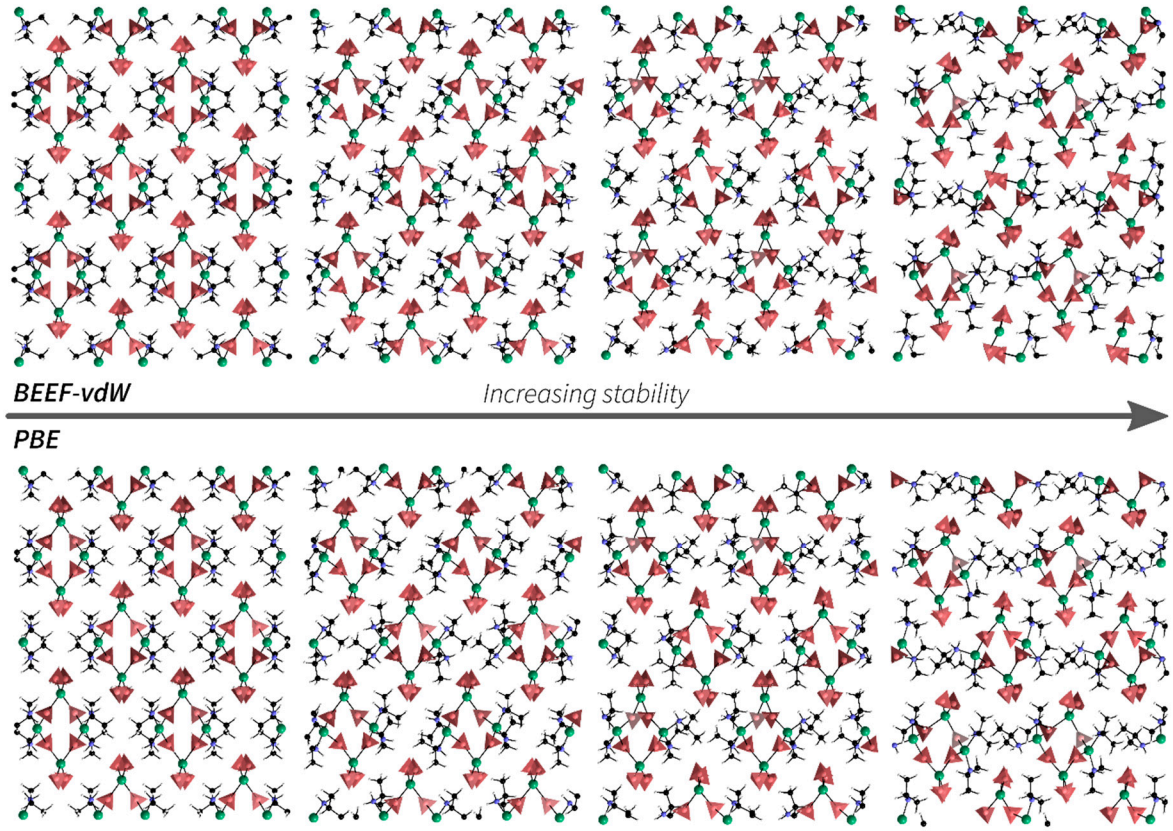

Figure S3. Structures gained from DFT in order of increasing stability using the PBE and BEEF-vdW functionals. Stability is increased as the 1-D helictical structure get more and more distorted. Differences between the functionals is mainly seen in the isopropyl groups, however the difference is minor. Due to the closeness of the isopropyl groups we expect that these contribute to the overall stability of the structure through weak dispersive interactions.

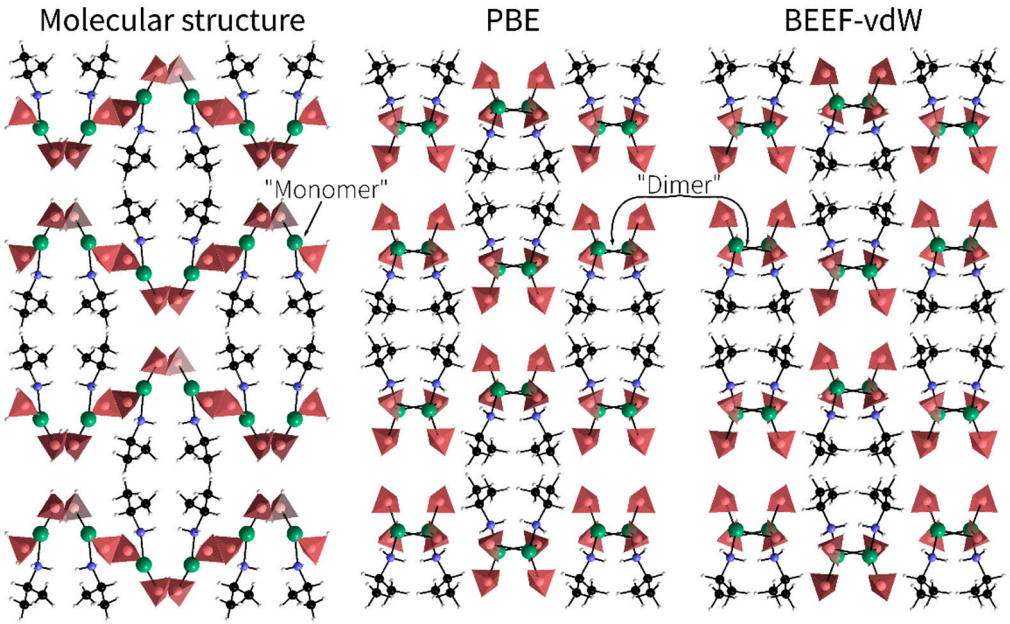

Figure S4. Rejected model found from ab inito structural solution. The structure was solved in C2, and upgraded to I222 with a good fit to the data. The structure consists of independent $Mg(BH_4)_2(CH_3)_2CHNH_2$ units meaning a maximum coordination number of 7 for

Mg, which is less than expected. The structure was found to be very unstable using both PBE and BEEF-vdw functionals. These resulted in the formation of a "dimer" between two Mg connected by two BH_4^-.

Table S1. DFT relative potential energies (in eV) for the ordered and increasingly disordered structures shown in Figure S3 and S4 calculated using the PBE and BEEF-vdW functionals. The energies are referenced to that of the ordered structure (defined as 0 eV). A more negative energy implies higher stability.

Structure / DFT functional	**PBE**	**BEEF-vdW**
Ordered	0	0
Disordered 1	−0.62	−0.61
Disordered 2	−0.70	−0.81
Disordered 3	−0.97	−1.35
I222_least	0.42	0.79

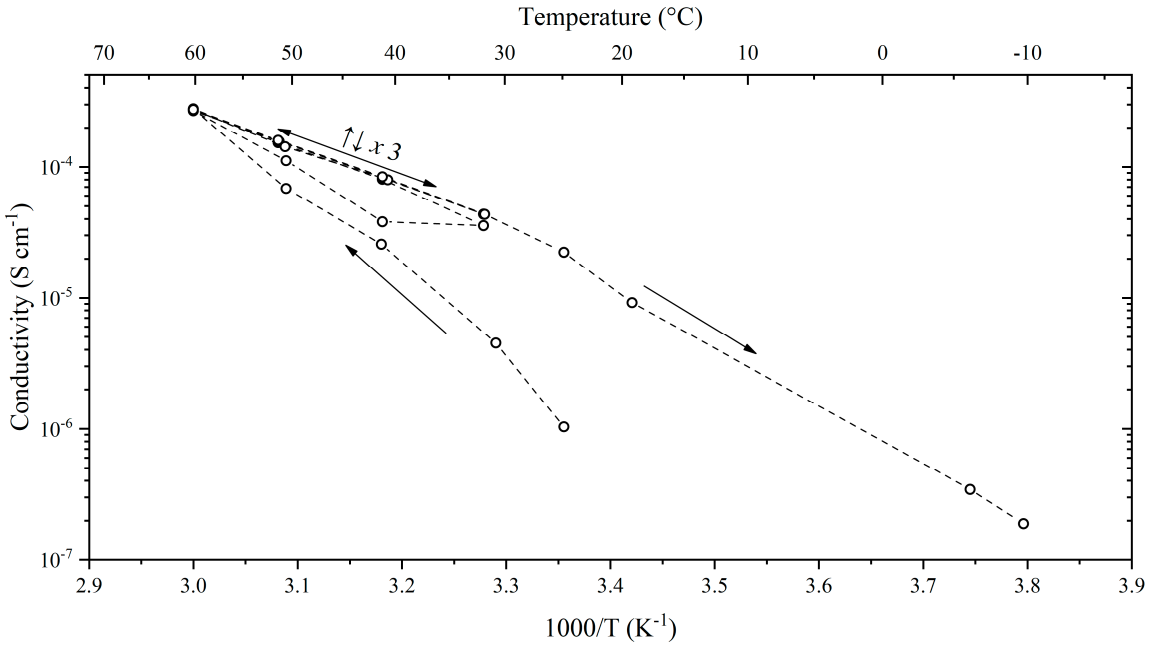

Figure S5. Ionic conductivity of $Mg(BH_4)_2$·IPA@Al_2O_3 (s5) without preheating. First a steep activation occurs, whereafter the sample gradually deforms during the 3 heating cycles. The highly conductive state is maintained during cooling to -10 ℃.

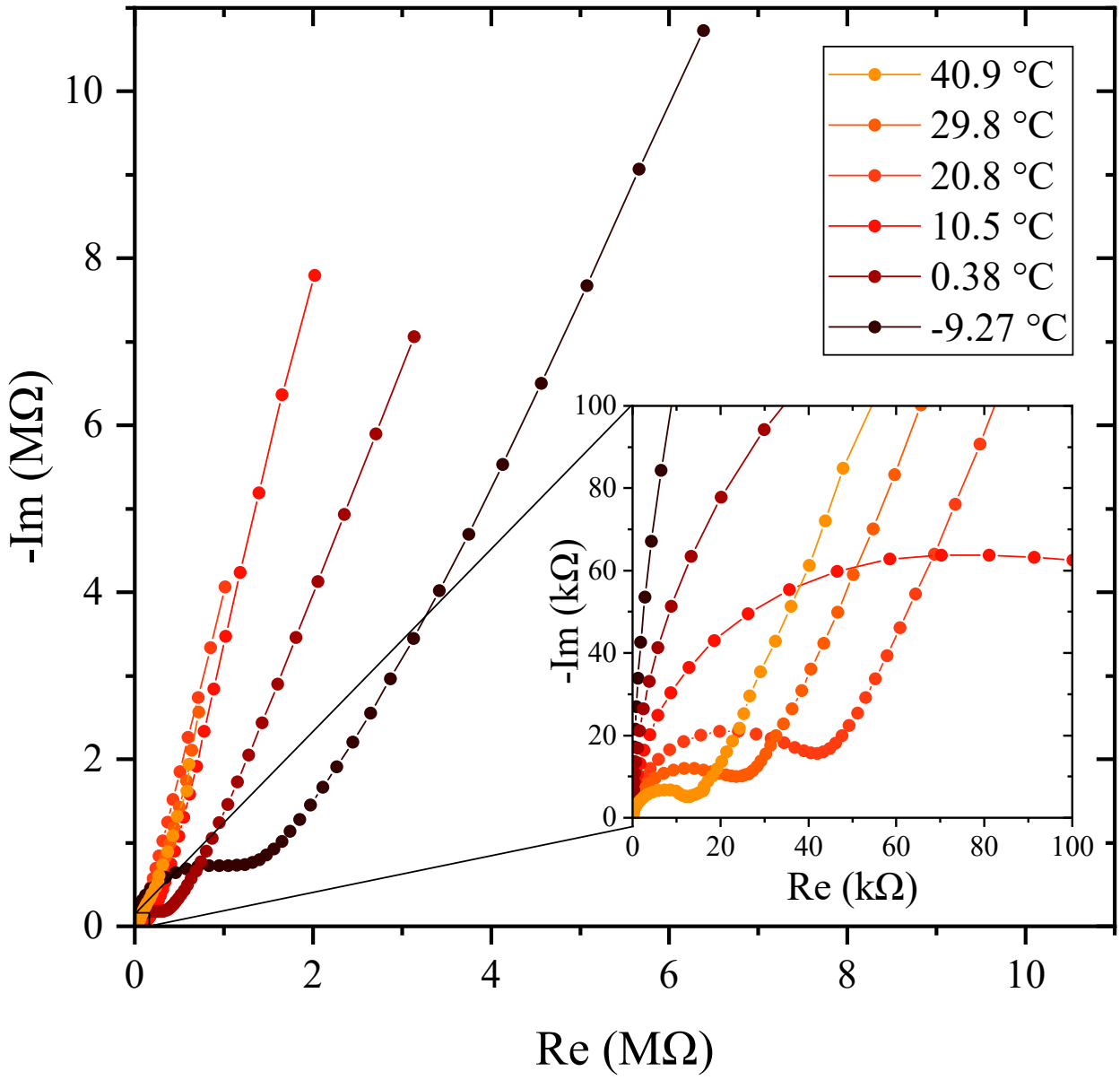

*Figure S6. Nyquist plot of $Mg(BH_4)_2$·1.2IPA-Al_2O_3 (**s5**) in the temperature range from -9.27 ℃ to 40.9 ℃. Inset shows a zoom in of the low resistance region. Measurements were performed from 7×10^7 Hz to 1 Hz.*