Electronic supplement

Table S1: theoretical and experimental vibrational frequencies of isolated water molecule

mode	theoretical frequency (this work) (cm^{-1})	theoretical frequency PAW[a] (cm^{-1})	exp. (observed) frequency[b] (cm^{-1})
symmetric stretching	3808	3836	3756
antisymmetric stretching	3696	3723	3657
bending	1596	1575	1595

cubic cell parameter: a=15.87 Å, single shifted k-point, d(OH)=0.96781Å, (HOH)=104.2735°
[a]T-Thienprasert, J., Boonchun, A., Reunchan, P., and Limpijumnong, S.: Identification of hydrogen defects in α-Al_2O_3 by first-principles local vibration mode calculations, Phys. Rev. B, 95, 134103, 2017.
[b]Shimanouchi, T., Tables of Molecular Vibrational Frequencies Consolidated Volume I, National Bureau of Standards, 1972, 1-160.

Table S2: Selected theoretical and experimental vibrational properties of diaspore:

mode	theoretical frequency (cm^{-1})	exp. (observed) frequency[a] (cm^{-1})
B1u bending	1016	964
B3u bending	1082	1078
B2u bending	1181	1153
B1u bending x 2	2032	1985
B3u bending x 2	2164	2116
B2u bending x 2	2362	2340
B2u stretching	2947	2920

orthorhombic cell parameters: a=4.426 Å b=9.489 Å c=2.866 Å, shifted 4x2x6 k-point grid, d(OH)=1.008 Å
[a]Delattre S., Balan E., Lazzeri M., Blanchard M., Guillaumet M., Beyssac O., Haussühl E., Winkler B., Salje E.K.H., Calas G. (2012) Experimental and theoretical study of the vibrational properties of diaspore (α-AlOOH) Phys. Chem. Minerals 39, 93-102.

The supplement contains:
- .cif structure files of the defective corundum models
- Normalized eigendisplacements of optical modes. Atoms are ordered as in the corresponding .cif structure file. Displacement coordinates are reported in a cartesian reference frame (X,Y,Z) defined such as the hexagonal lattice vectors are a=(1,0,0), b=(-1/2, sqrt(3)/2,0) c=(0,0,1).
- .mov files with animations showing the 4 relevant modes (stretching, 2 bending, coupled mode) for the $(H^+)_i$ model.
- Spectra from the GIA database as .txt files, in three folders containing spectra associated with '3161' series, Mg "Punsiri" stones and Ni-doped synthetics.