# Supporting Information for

# "Magnetic anisotropy on demand exploiting high-pressure as remote control: an *ab initio* proof of concept"

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| Press. (GPa) | Volume (Å³) | a (Å) | b (Å) | c (Å) | α (°) | β (°) | γ (°) |
|---|---|---|---|---|---|---|---|
| Exp | 1213.5(2) | 8.7190(10) | 9.0790(10) | 15.6370(10) | 82.962(5) | 85.824(5) | 81.637(5) |
| 0 | 1.198.201 | 8.509 | 9.182 | 15.623 | 82.905 | 85.507 | 82.425 |
| 3 | 1.063.343 | 8.143 | 8.765 | 15.192 | 82.620 | 84.991 | 82.529 |
| 4 | 1.031.624 | 8.083 | 8.619 | 15.117 | 82.346 | 84.455 | 82.573 |
| 5 | 1.009.649 | 8.056 | 8.472 | 15.125 | 81.722 | 84.143 | 82.736 |
| 5.5 | 1.000.697 | 8.038 | 8.438 | 15.093 | 81.602 | 84.051 | 82.707 |
| 5.6 | 1.002.171 | 8.044 | 8.442 | 15.094 | 81.626 | 84.065 | 82.735 |
| 5.7 | 993.282 | 8.080 | 8.361 | 15.078 | 81.172 | 83.486 | 82.438 |
| 5.8 | 985.007 | 8.047 | 8.347 | 15.033 | 81.205 | 83.214 | 82.858 |
| 6 | 986.784 | 8.042 | 8.373 | 15.007 | 81.458 | 83.450 | 82.792 |
| 7 | 963.666 | 7.984 | 8.260 | 14.958 | 81.706 | 82.876 | 83.323 |
| 9 | 933.267 | 7.958 | 8.184 | 14.707 | 81.522 | 82.279 | 82.744 |
| 10 | 920.524 | 7.913 | 8.148 | 14.648 | 81.563 | 82.213 | 82.939 |
| 12 | 893.335 | 7.853 | 8.036 | 14.540 | 81.457 | 81.763 | 83.056 |

**Table S1**. Experimental and optimized cell parameters at several pressures.
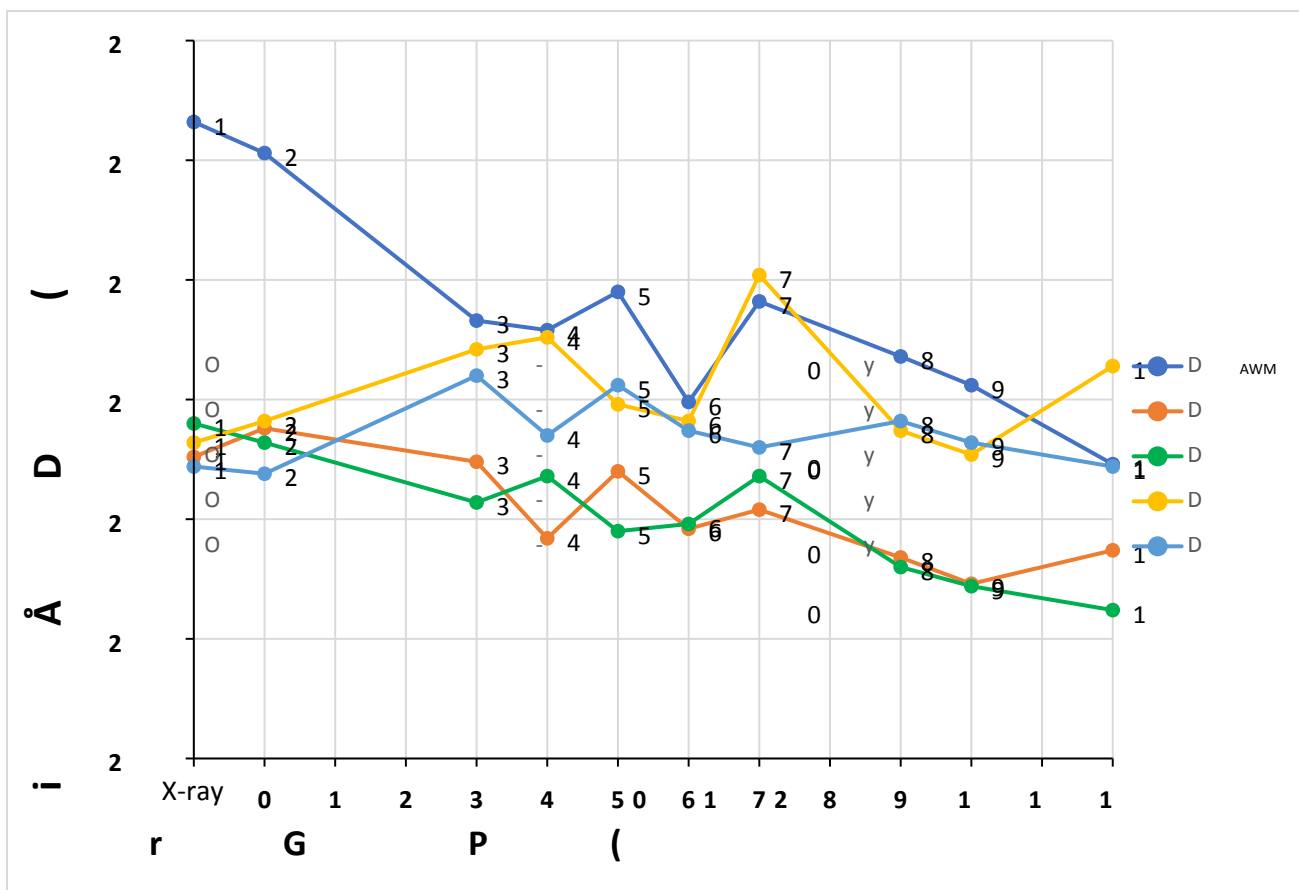
**Figure S1.** Bond distances between dysprosium and the five oxygen atoms in the optimized structure at different pressures of **1**.
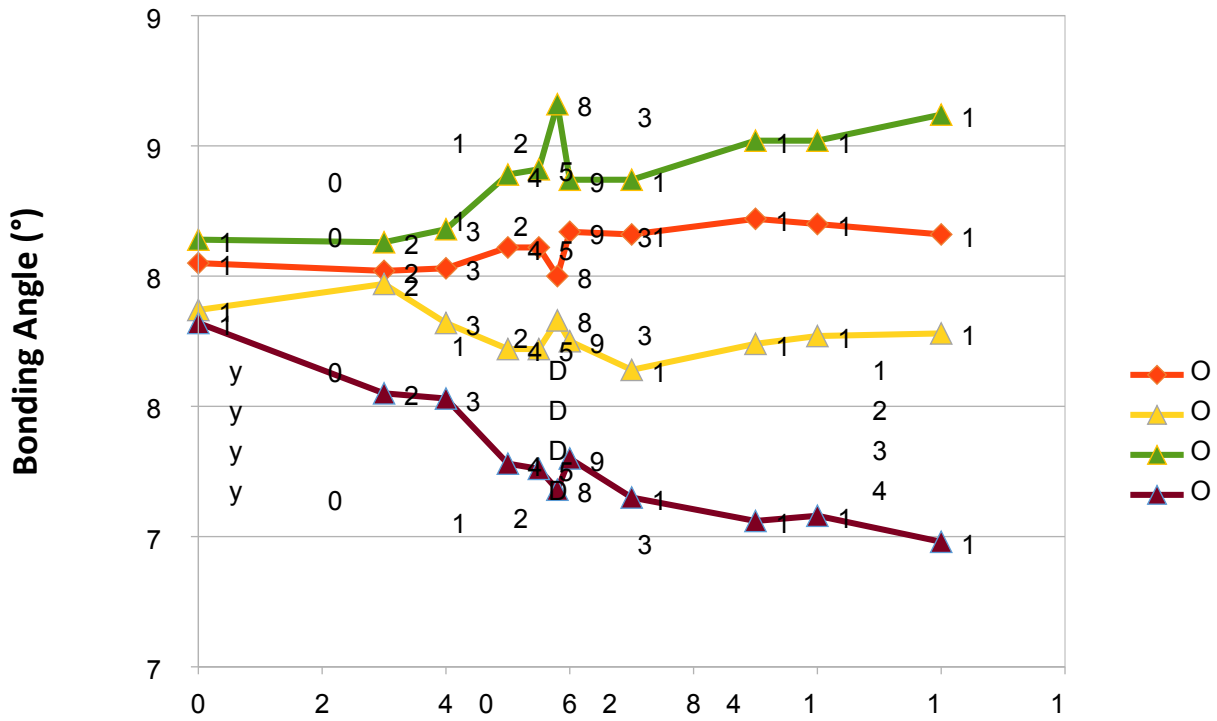
**Figure S2.** Bond angles between dysprosium and the four oxygen atoms in the optimized structure at different pressures of **1**.
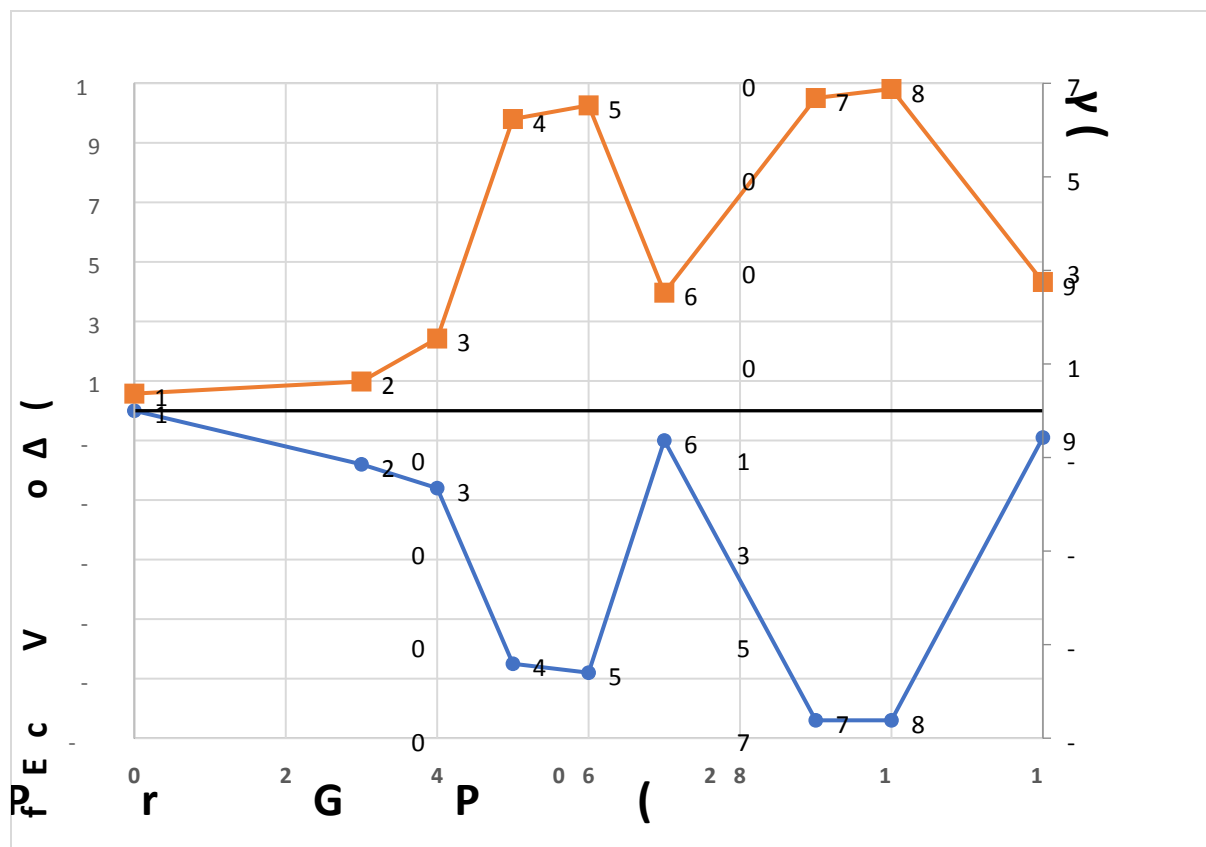
**Figure S3.** Model **M2m**. Variation of the energy gap between ground and first excited KD with respect to the value computed at 0 GPa (Blue) and angle between computed and experimental MEA (orange).
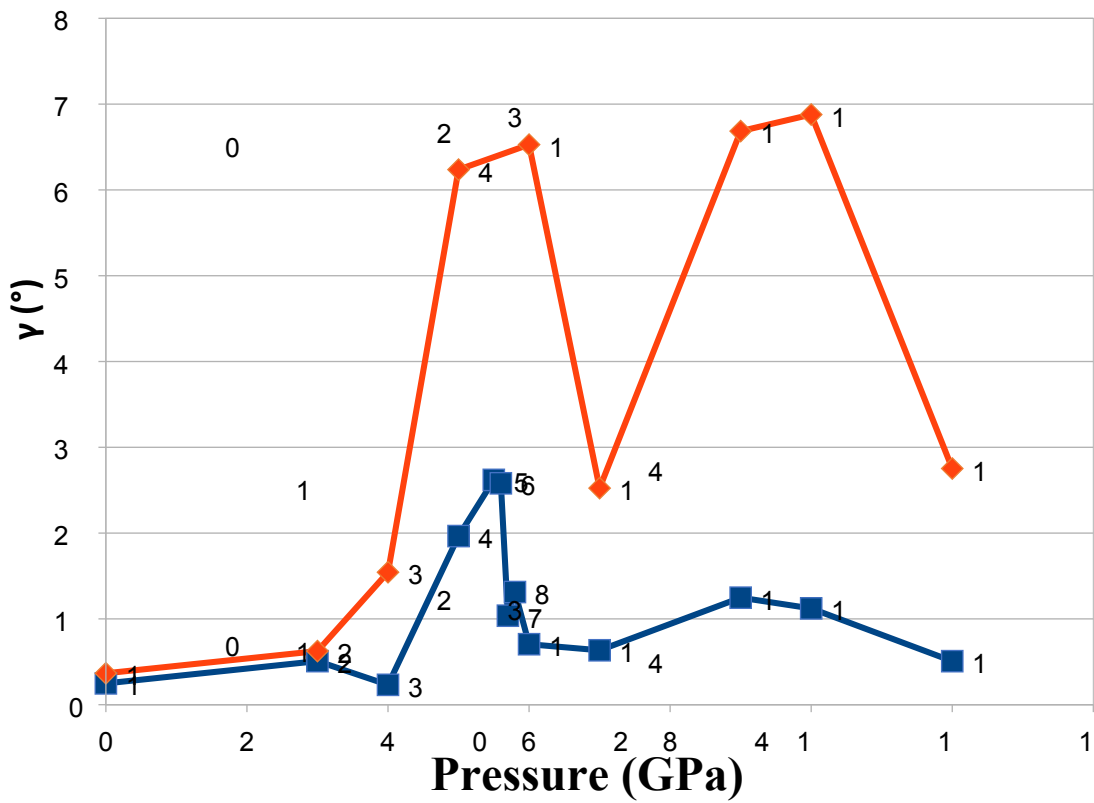
**Figure S4.** Angle between the computed magnetic easy axes and the experimental one at standard pressure (1 bar) of the ground KD for models **M1** (blue) and **M2m** (orange), for several values of the simulated pressure.
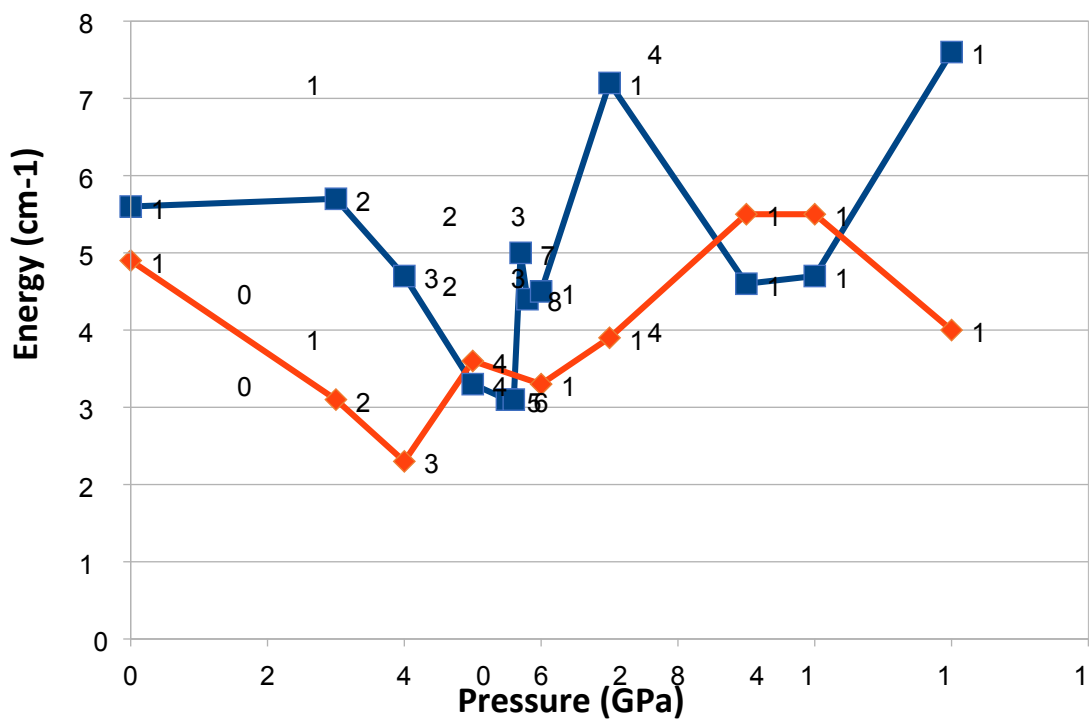
**Figure S5.** Energy of the first excited KD for models **M1** (blue) and **M2m** (orange), computed on the optimized geometry for several values of the external pressure