Supplementary Material:
Pressure effect on hydrogen tunneling and vibrational spectrum in α-Mn

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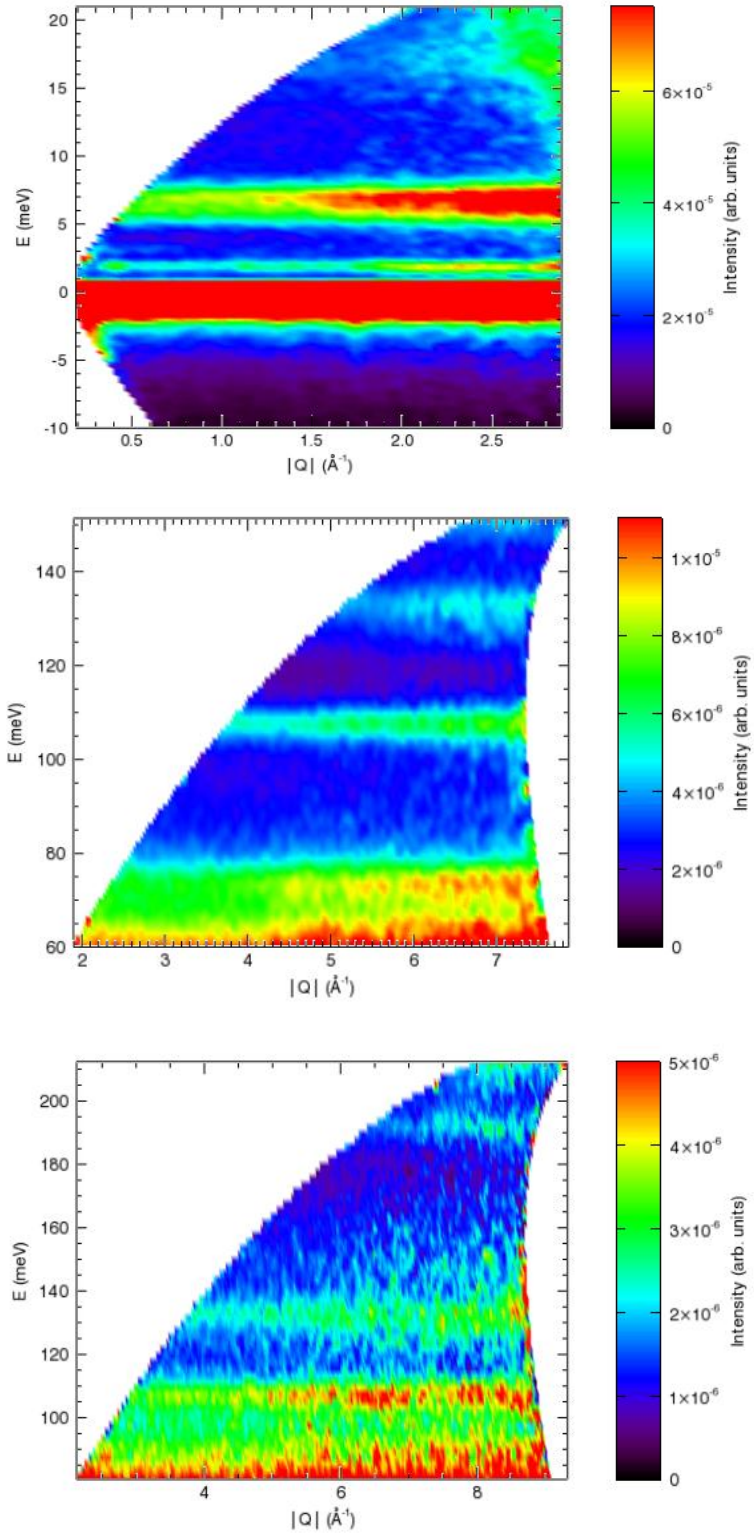

FIG. S1: *(Q,E)* contour plots of *S(Q,E)* spectra of α-$MnH_{0.07}$ measured at a 22 kbar pressure and *T*=6 K using the SEQUOIA spectrometer with E_i=25, 160 and 225 meV shown in the top, middle and bottom pictures, respectively.

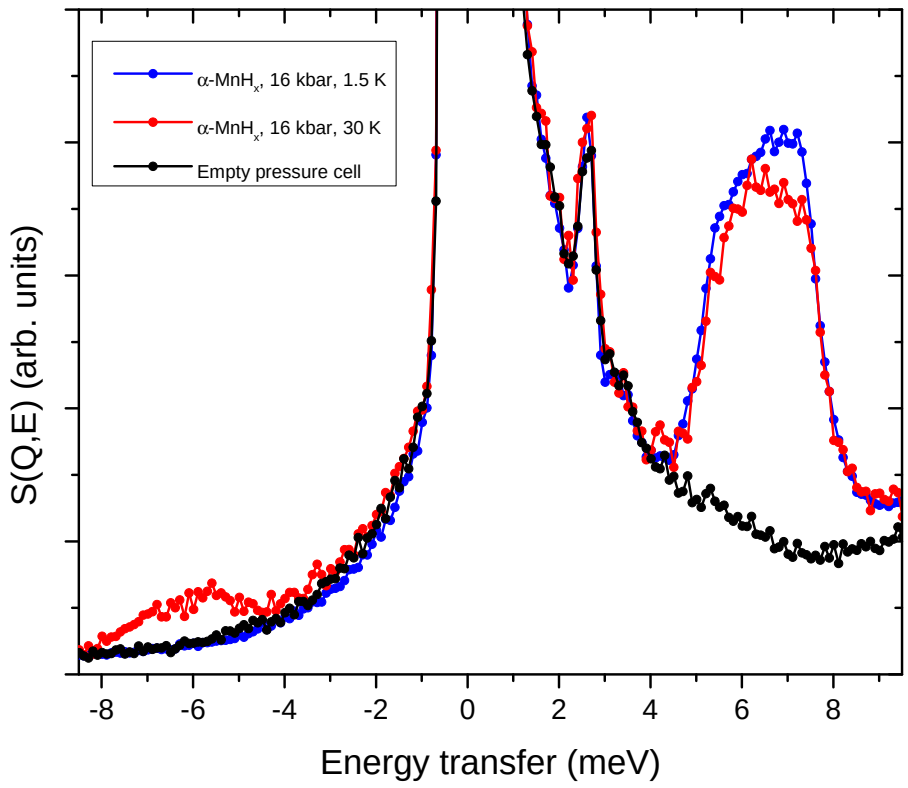

FIG. S2: Raw INS spectra of α-$MnH_{0.07}$ measured at a 16 kbar pressure at T=1.5 K and 30 K using the CNCS spectrometer with E_i=12 meV. The black curve shows the spectrum of the pressure-cell with fluorinert in the cryostat at ambient pressure and T=1.5 K. The peak at ~2.5 meV is a spurious peak from the pressure-cell; overall the spectra show a good signal (tunneling peak between 5 and 8 meV) to background ratio. The leftmost peak (between -5 and -8 meV, on the neutron energy gain side) in the spectrum of α-$MnH_{0.07}$ at T=30 K is due to transition from the upper to lower split ground states of tunneling hydrogen (the upper state is temperature populated at 30 K).

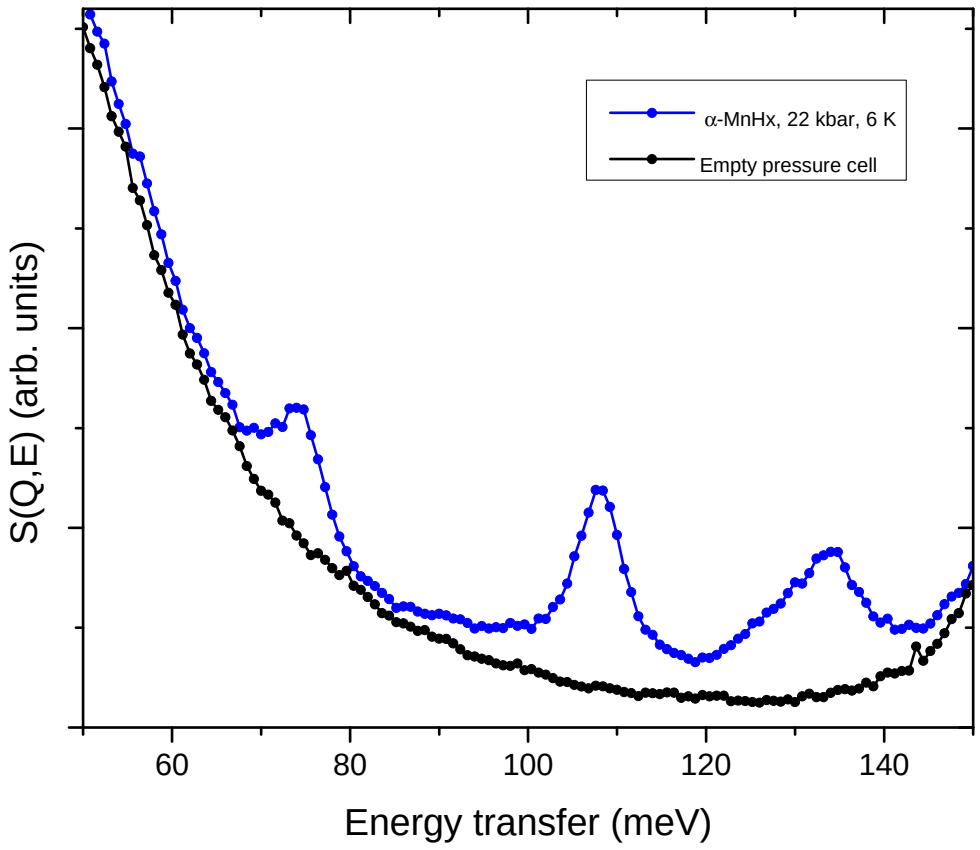

FIG. S3: Raw INS spectrum of α-$MnH_{0.07}$ measured at a 22 kbar pressure and T=6 K using the SEQUOIA spectrometer with E_i=160 meV. The black curve shows the spectrum of the pressure-cell with fluorinert at ambient pressure and T=6 K. The data show a good signal to background ratio in the energy range of interest (hydrogen optical modes at about 73, 106 and 131 meV).

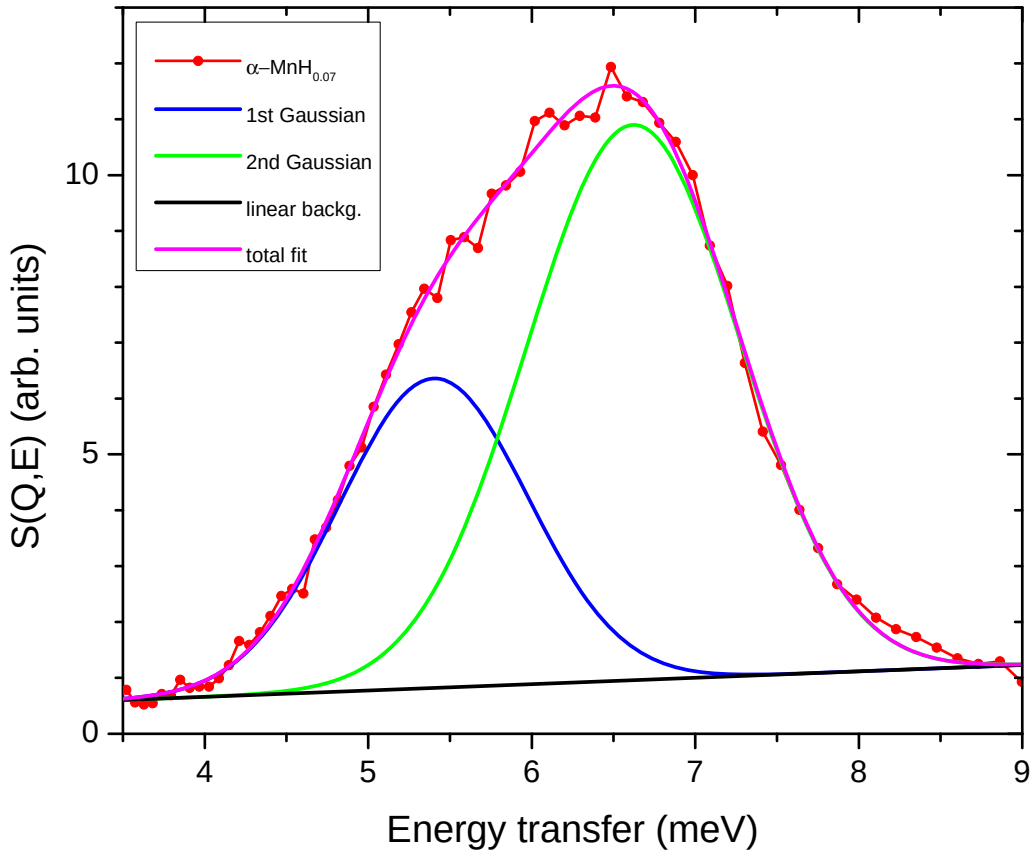

FIG. S4: INS spectrum of α-$MnH_{0.07}$ measured at 23 K and ambient pressure (without a pressure cell) using the TFXA spectrometer at ISIS (the data are taken from Ref. 13). The curve with the red points shows the experimental spectrum; the pink solid line is a fit with the two Gaussian functions and the sloped liner background, which are shown separately as the blue, green and black curves, respectively. The ratio of the areas under the 2^{nd} Gaussian and the 1^{st} Gaussian is about 2, which agrees with the supposed tetragonal distortion in the arrangement of the Mn atoms on the $24g_2$ sites (the nearest neighbors of the hydrogen atom) in the antiferromagnetic state.