Supplemental Material:

From parallel to single crystallization kinetics in high-density amorphous ice

Markus Seidl,[a] Katrin Amann-Winkel,[a] Philip H. Handle,[a] Gerhard Zifferer,[b]
Thomas Loerting[a,*]

[a]Institute of Physical Chemistry, University of Innsbruck, Innrain 52a, A-6020 Innsbruck,
Austria
[b]Department of Physical Chemistry, University of Vienna, Währinger Str. 42, A-1090 Wien,
Austria

e-mail: thomas.loerting@uibk.ac.at
phone: 0043/512/507-5062
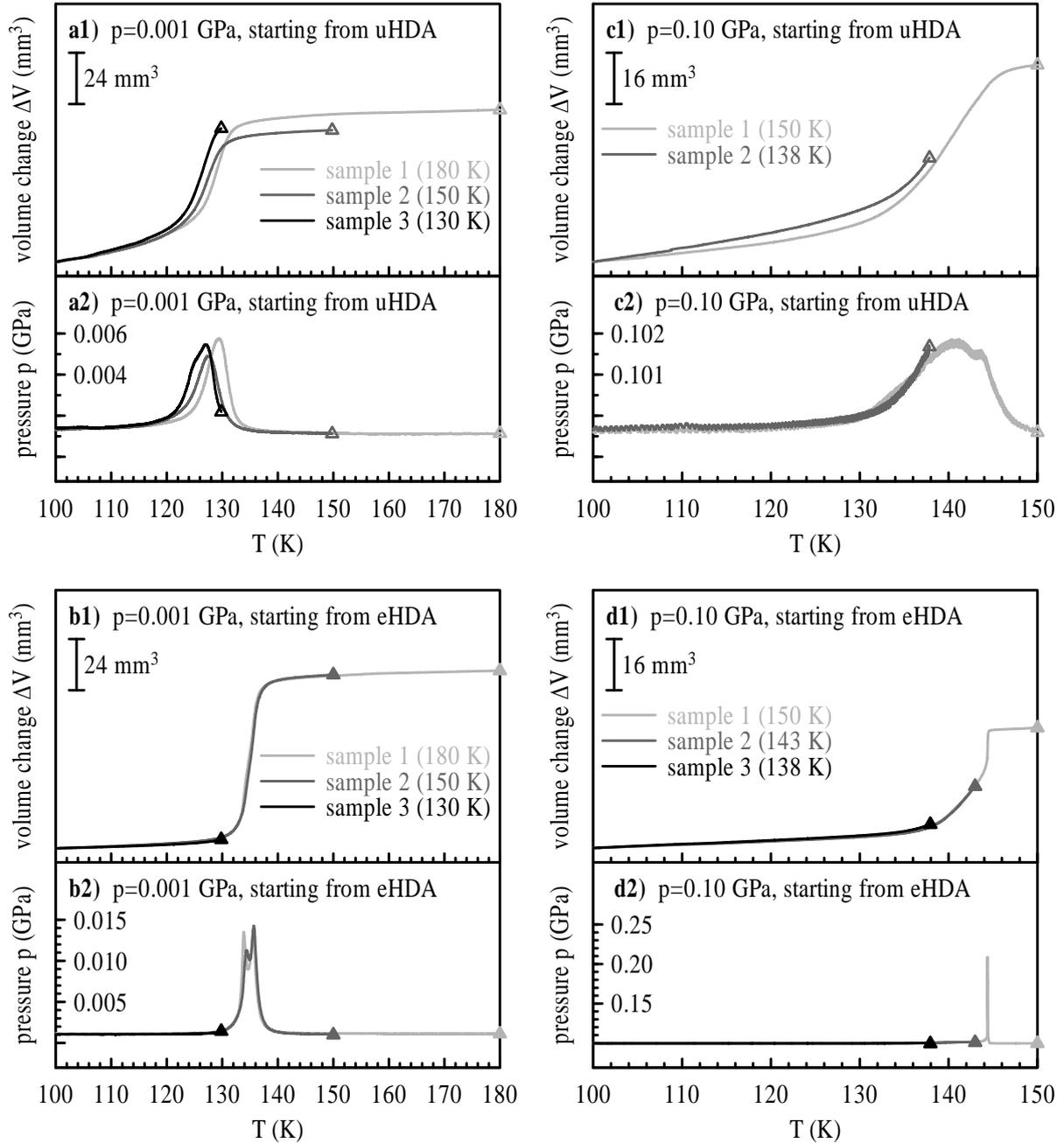
FIG. SM1. Reproducibility of the volume change and pressure evolution during isobaric heating of high-density amorphous ices at 0.001 GPa (left column) and 0.10 GPa (right column). The two or three distinct samples were prepared to gain powder diffractograms for states reached upon heating to different maximum temperatures (see Fig. 2, bottom panels). Triangles mark the end points of the curves, i.e. they indicate the states at the respective maximum temperatures (which are numerically given in the legend). In some cases the actual pressure significantly deviates from the nominal pressure in the temperature range where phase transformations take place (a2, b2 and d2). However, the phase behavior is governed by the pressure at the very onset of the transformation and, thus, the deviations at higher temperatures have no qualitative impact on the results.
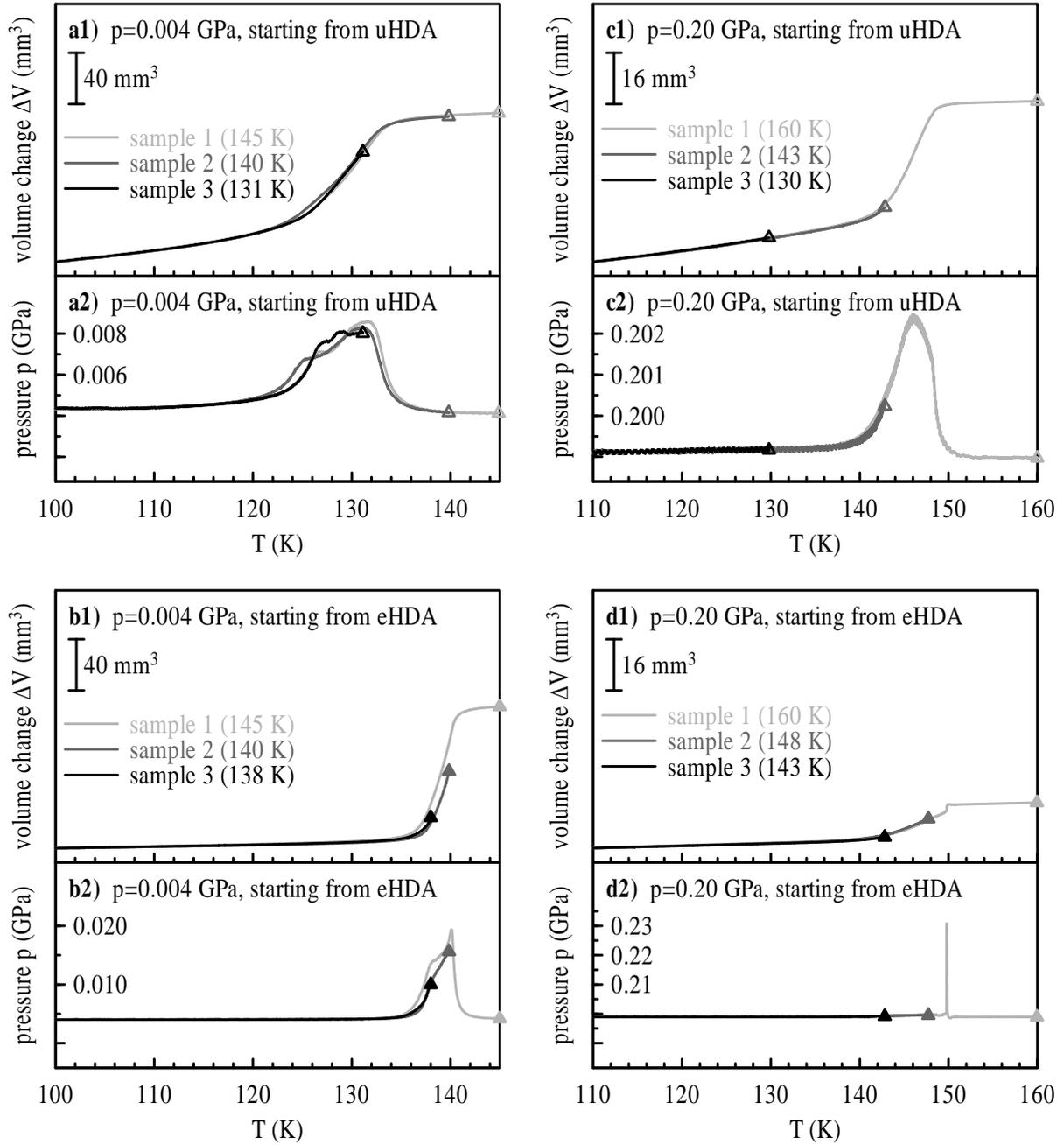
FIG. SM2. Reproducibility of the volume change and pressure evolution during isobaric heating of high-density amorphous ices at 0.004 GPa (left column) and 0.20 GPa (right column). The three distinct samples were prepared to gain powder diffractograms for states reached upon heating to different maximum temperatures (see Fig. SM2, bottom panels). Triangles mark the end points of the curves, i.e. they indicate the states at the respective maximum temperatures (which are numerically given in the legend). In some cases the actual pressure significantly deviates from the nominal pressure in the temperature range where phase transformations take place (a2 and b2). However, the phase behavior is governed by the pressure at the very onset of the transformation and, thus, the deviations at higher temperatures have no qualitative impact on the results.
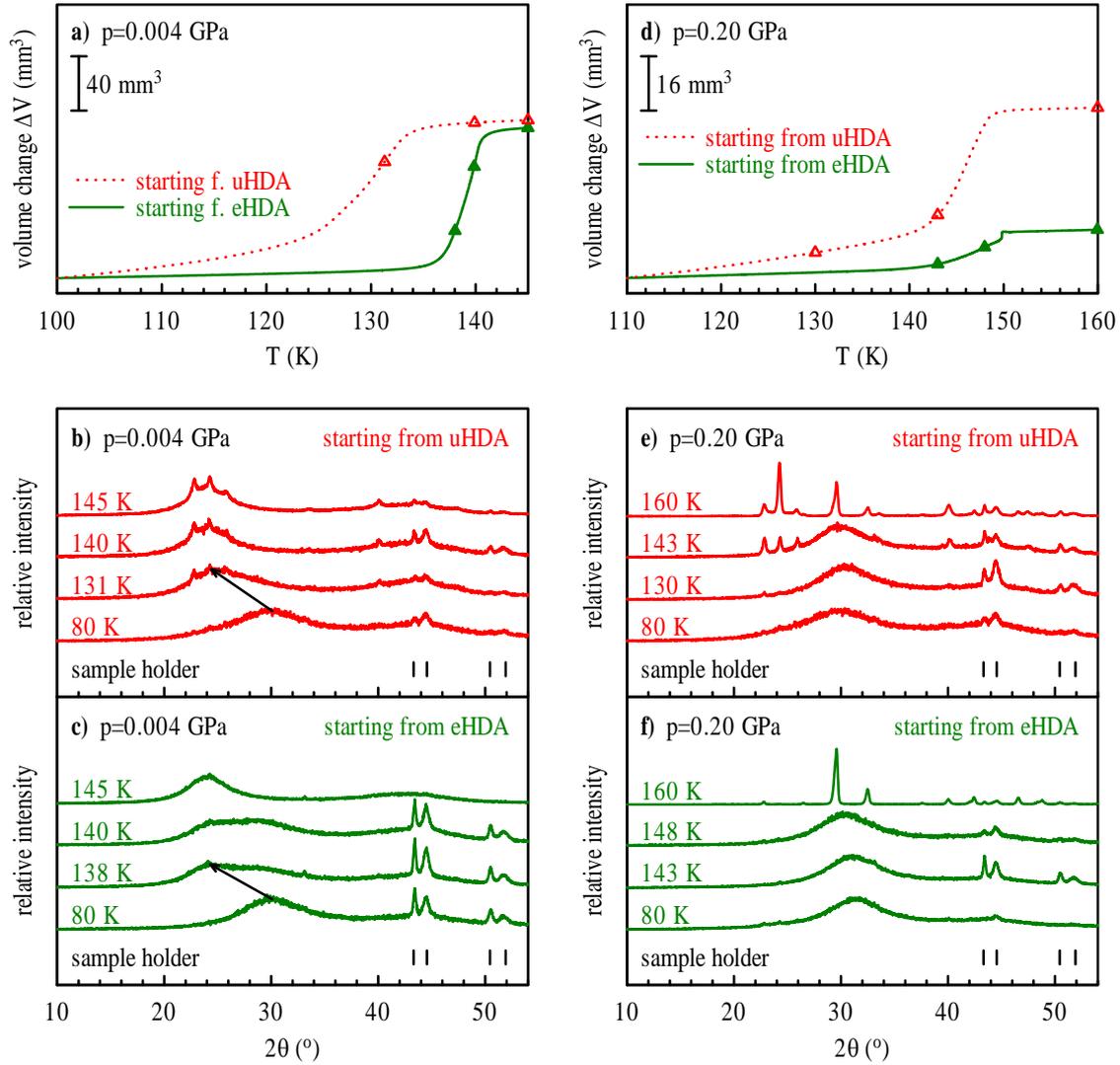
FIG. SM3. Isobaric crystallization of high-density amorphous ice. Volume-temperature curves upon isobaric heating of uHDA (red dotted curve) and eHDA (green solid curve) are shown for 0.004 GPa and 3 K/min (a) and 0.20 GPa and 2 K/min (d). Triangles indicate the states for which powder diffractograms have been recorded. Corresponding diffractograms for samples recovered to ~80 K and (sub)ambient pressure are shown for both starting material uHDA (b, e) and eHDA (c, f). Curves are offset for clarity. Temperature labels indicate the maximum temperature experienced in isobaric heating runs and arrows mark the shift of halo peak maxima reflecting the transformation of HDA to LDA. The starting material uHDA is characterized by a broad halo peak with a maximum at ~30.1° (panels (b) and (e)), while the starting material eHDA shows a peak maximum at ~30.1° (panel (c)) and ~31.4° (panel (f)), respectively. The phase composition reads as follows: (b) HDA (80 K), LDA/HDA/ice Ih (131 K), LDA/ice Ih (140–145 K); (c) HDA (80K), HDA/LDA (138–140 K), LDA (145 K); (e) HDA (80 K), HDA/ice Ih (143 K), ice IX/ice I (160K); (f) HDA (80–148 K), ice IX (160 K). In the case of 0.004 GPa uHDA was prepared by compression of ice Ih to 1.7 GPa at ~80 K and eHDA by decompression of VHDA to 0.10 GPa at 140 K. In the case of 0.20 GPa uHDA was prepared by compression of ice Ih to 1.6 GPa at ~80 K and eHDA by decompression of VHDA to 0.20 GPa at 140 K.