

Supporting information

Pressure-induced reversible phase transitions in a new metastable phase of vanadium dioxide

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Synthesis of VO₂ (Mx') under Pressure

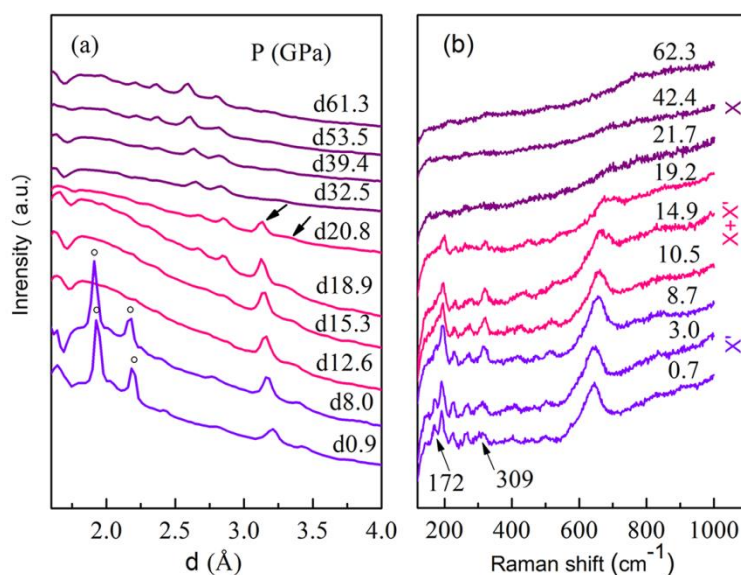


Figure S1. High-pressure (a) X-ray diffraction patterns and (b) Raman spectra of VO₂ collected upon decompression. Circles indicate diffraction peaks from Fe.

Figure S1 show the X-ray diffraction patterns and Raman spectra for VO₂ collected upon decompressed from about 61 GPa. In figure 1a, all diffraction peaks shift to larger d-spacing without any modification of the overall peak pattern above 32.5 GPa. When pressure decreased down to 20.8 GPa, new peaks at about 3.1 Å and 3.3 Å appear. This indicates that the structural transition, from the high-pressure X to a new phase (named Mx'), takes place at about 20.8 GPa. With further decreasing pressure, all the diffraction peaks of X phase gradually weakens and finally disappear at ~12.6 GPa, suggesting that all the sample transformed into the Mx'. Additional evidence of this structural phase transition is shown in Raman measurements. In figure 1b shows the selected Raman spectra measured upon releasing from 62 GPa. With decreasing pressure, we did not observe any Raman peak above 19.2 GPa, because this pressure regime (62~19 GPa)

corresponds to the stable pressure range of the metallic X phase, for which the Raman signals are too weak to detect. Upon further decompressing, Raman peaks start to appear. The intensities of these Raman peaks increase with decreasing pressure, and the Raman peak at about 195 cm^{-1} gradually broadens and finally splits into two peaks below 10.5 GPa, indicating that all the VO_2 samples in the X phase have transformed into the new monoclinic Mx' phase.

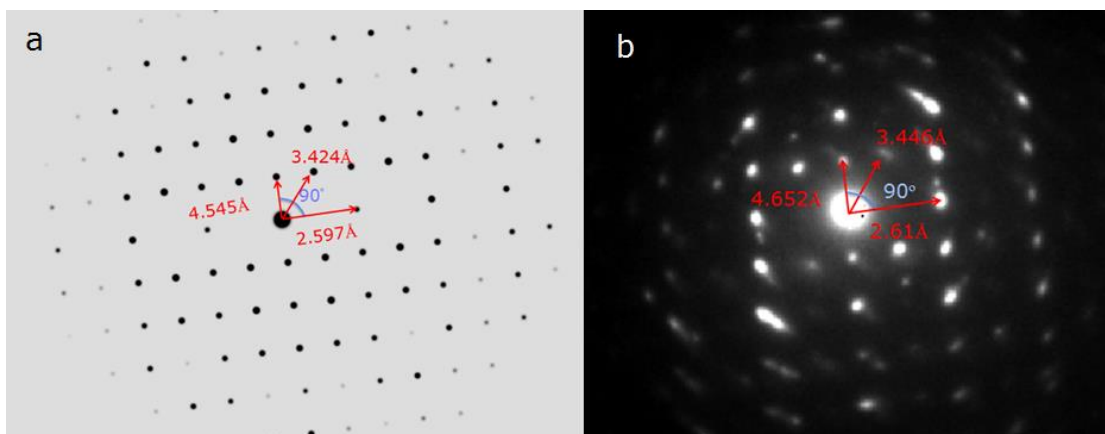


Figure S2. (a) The simulated the TEM/SAED patterns, (b) Observed diffraction pattern.

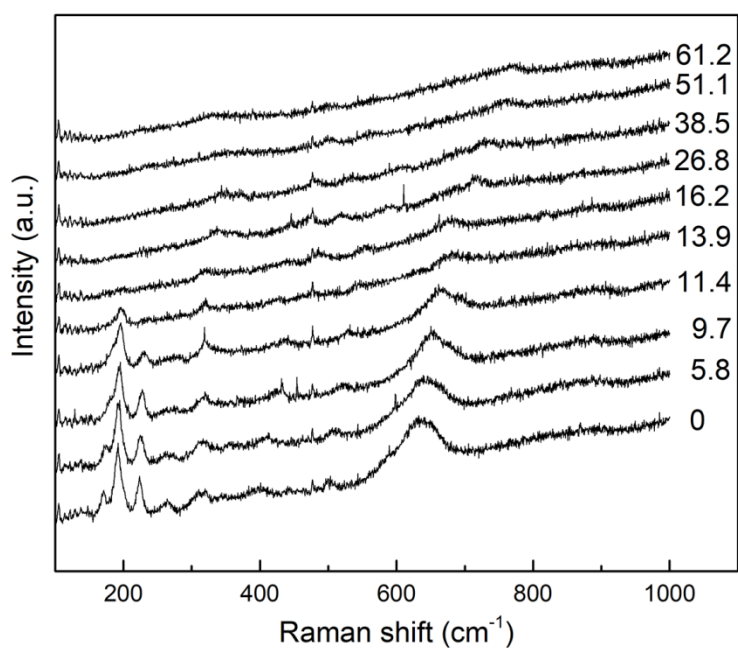


Figure S3. Raman spectra collected upon decompression.