

Ordering and elasticity associated with low-temperature phase transitions in lawsonite

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ABSTRACT

The two low-temperature phase transitions of lawsonite have been studied using single-crystal X-ray diffraction from 86 to 318 K and a single-crystal high-frequency continuous-wave resonance technique from 323 to 102 K. While recently published data of the variations of strains, birefringence, and IR line widths are consistent with the (271 K) *Cmcm-Pmcn* transition being simply tricritical, our investigation of critical X-ray reflections and the six diagonal elastic constants of lawsonite reveals, consistently, a more complex crossover pattern in the temperature range of 205–225 K. Below 205 K the overall pattern is again in good agreement with a tricritical solution of the *Cmcm-Pmcn* transition and a second-order behavior of the (120 K) *Pmcn-P2₁cn* transition. The structure determination from single-crystal X-ray data at 215 K reveals a possible orientational disorder of some of the hydroxyl groups in the *Pmcn* phase. From this and a recent strain analysis of deuterated and hydrogenated lawsonite we conclude that down to 205 K the *Cmcm-Pmcn* transition is driven by a displacive component, as observed in strain and birefringence data, plus an order/disorder component or dynamical effects associated with proton ordering. Below 205 K only the displacive component plays a role, and the (120 K) *Pmcn-P2₁cn* transition is driven by a single order parameter. The remarkable elastic softening of C_{66} ahead of the *Cmcm-Pmcn* transition indicates another orthorhombic-monoclinic transition, which is suppressed on cooling through the low-temperature phase sequence *Cmcm-Pmcn-P2₁cn*, but can be observed on applying pressure to the mineral.