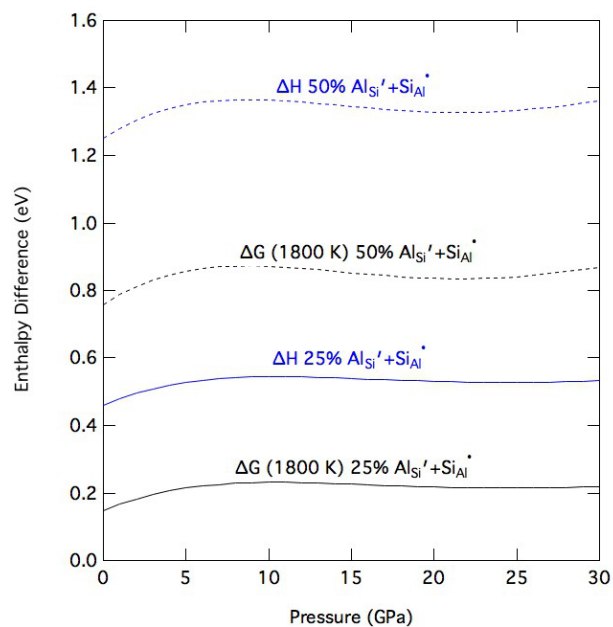**SUPPLEMENTAL FIGURE S1.** Plot of total energy as a function of cut off energy (red symbols). Please refer to the left and lower axis. Plot of total energy as a function of irreducible **k**-points (blue symbols). Please refer to the top and right axis.

SUPPLEMENTAL FIGURE S2. The enthalpy of disorder (blue) per molecular unit for calculations with 25% (solid) and 50% (dashed) $Al_{Si'} + Si_{Al*}$ coupled substitutions in phase egg. The Gibbs Free Energy difference (black) at 1800 K is calculated assuming that the contribution to entropy difference between the ordered and disordered is configurational.
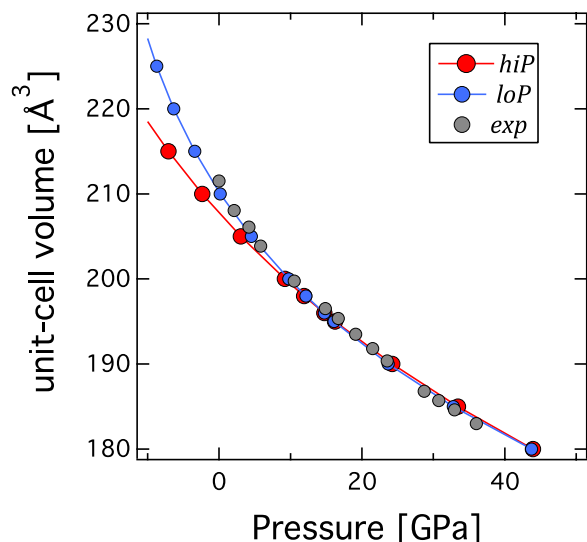
SUPPLEMENTAL FIGURE S3. Plot of unit-cell volume vs. pressure. Dark blue symbols represent phase egg (*loP*), red symbols represent phase egg (*hiP*), and gray filled symbols represent experimental results. This is typical behavior for a second order transition where the high pressure and the low pressure phase converge asymptotically at pressures greater than the transition pressure.
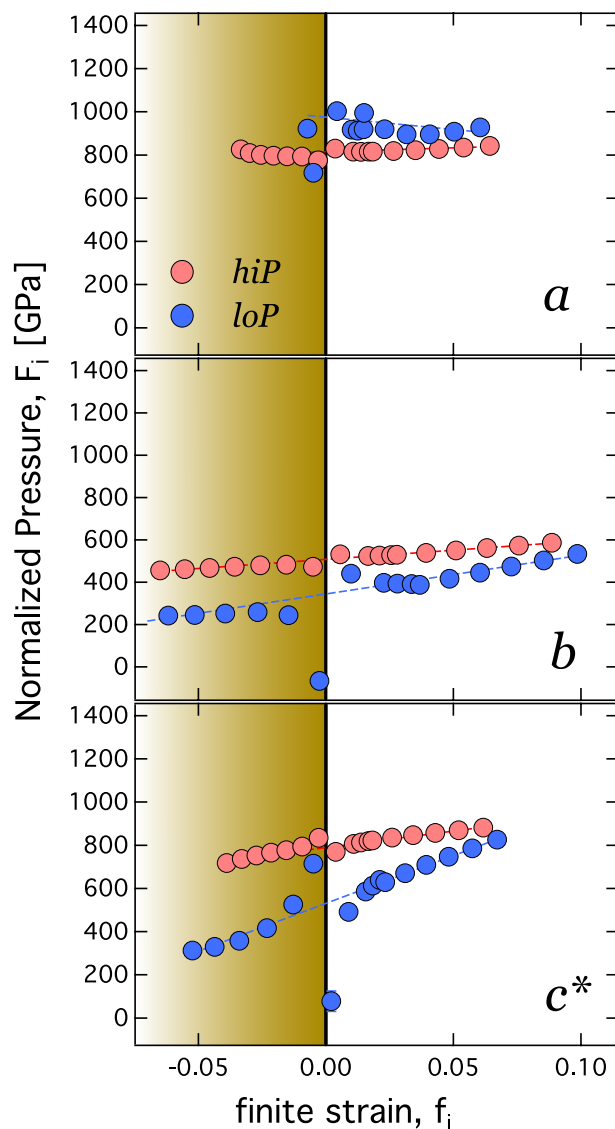
SUPPLEMENTAL FIGURE S4. Plot of linear normalized pressure (F_l) vs. Eulerian linear finite strain (f_l) for (**a**) *a*-, (**b**) *b*-, and (**c**) *c**-axis directions. Dark blue symbols phase egg (*loP*) and red symbols phase egg (*hiP*). The linear compressibility, K_l related to the normalized pressure and finite strain with the expression: $F_l = K_l + m_l f_l$; where is the linear normalized pressure, given by $F_l = P/[f_l(1 + 2f_l)(1 + 2f_V)]$ and f_l is the linear Eulerian finite strain, given by $f_l = \frac{1}{2}[(l_0/l)^2 - 1]$ and f_V is the volume Eulerian finite strain given in Equation 2 of the main text.