Soft branches, central peak, and strong isotropic negative thermal expansion in a perovskite material

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Large, isotropic negative thermal expansion is known to exist in only a handful of materials, beginning with the discovery of ZrW_2O_8 in the 1990s. In 2010, perovskite fluoride ScF_3 was discovered to have a similarly profound negative thermal expansion (NTE) effect, shrinking in response to heat over a 1000 K temperature window with a linear thermal expansion coefficient lower than -10^{-5}/K. Another curious property of this material is the structural stability – ScF_3 retains a simple cubic structure and four atom unit cell from cryogenic temperature to its high melting point of 1800 K.

We present a high energy resolution inelastic X-ray scattering study of single crystalline ScF_3 in order to examine the anharmonic phonon dynamics that underpin the NTE behavior. Surprisingly, we find that an entire optical mode branch circumscribing the Brillouin zone boundary softens to nearly zero frequency as the temperature T approaches T=0. ScF_3 at T=0 thus sits in extreme proximity to a quantum phase transition. We interpret this result in the context of better studied trifluorides and examine in detail the disorder phase diagram. In addition, concomitant with softening of the optic branch, a "quantum central peak" emerges and strengthens for $T<100$ K, which appears to steal and exhaust spectral weight from the optic mode. These extraordinary observations give insight into the central peak phenomena and may have implications to other perovskite-structured materials.

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