# Archetypal soft-mode driven antipolar transition in francisite $Cu_3Bi(SeO_3)_2O_2Cl$
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# Supplemental Material

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## I. PHONON DISPERSION BY INELASTIC X-RAY SCATTERING

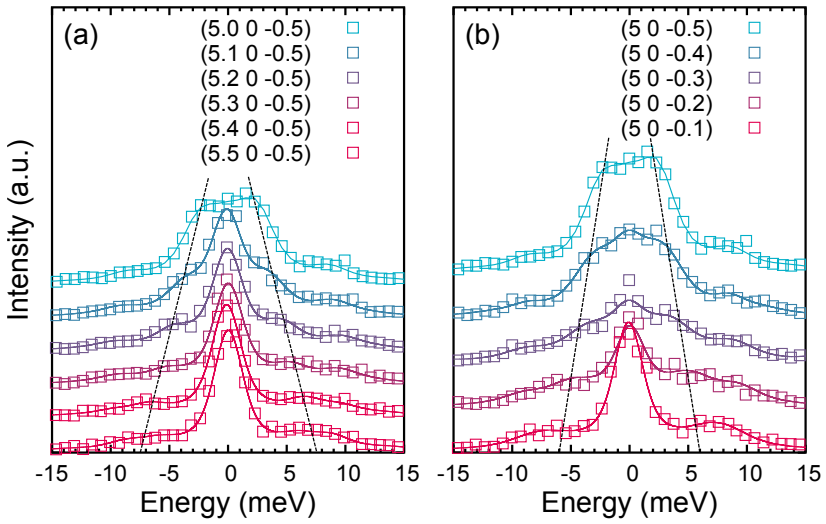


Figure 1: Dispersion (a) along the $h$ direction and (b) along the $l$ direction measured at room temperature. The solid line is the result from the fit and the dotted line is a guide for the eye following the soft mode.

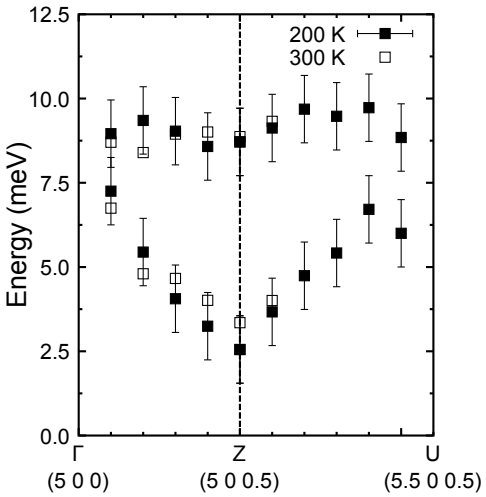


Figure 2: Dispersion along the Γ–$Z$–$U$ path at 200 and 300 K measured by Inelastic X-Ray Scattering.

## II. CALCULATION DETAILS

The calculations were carried out within the Density Functional Theory framework as implemented in the VASP code, using the Perdew-Burke-Ernzerhof approximation to the exchange-correlation functional. The strong electronic correlations were taken into account within the GGA+U approach. The Coulomb repulsion was set to U=9.5 eV for the d orbitals of Cu, while the exchange coupling was set to J=1.0 eV. We employed a plane-wave energy cutoff 500 eV for the basis set in all our calculations. A 3×2×3 (3×2×1) $k$-mesh was used in the calculations of the $Pmmn$ ($Pcmn$) primitive cell. Atoms were allowed to relax until atomic forces became smaller than 0.01 eV/Å. The phonon dispersions were calculated using the frozen-phonon approach in 120-atom supercells (2×1×2) supercells with respect to the $Pmmn$ primitive cell).

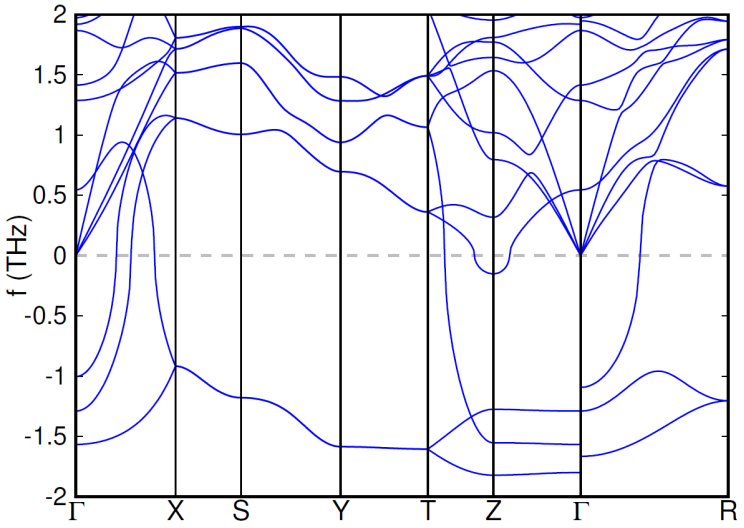


Figure 3: Phonon dispersion of Francisite in the high symmetry phase ($Pmmn$) as computed in its primitive cell.

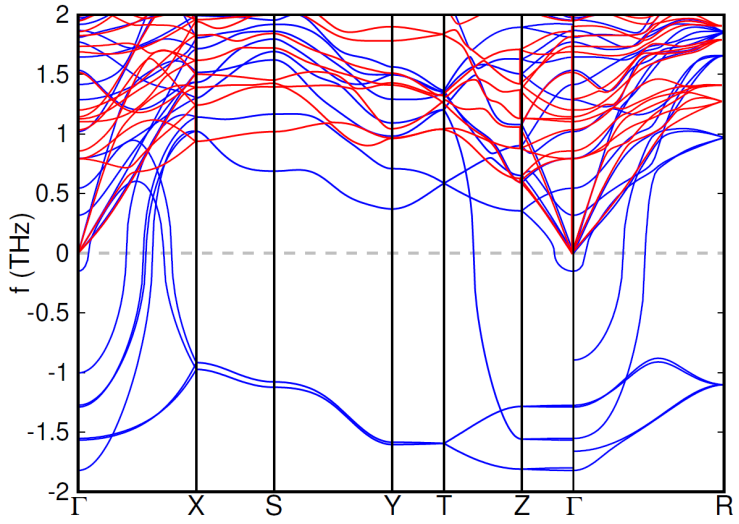


Figure 4: Phonon dispersion of Francisite in the high symmetry phase (blue) and low symmetry (red) phases. The former has now been computed in a 1x1x2 supercell with respect to its primitive cell, so that it can be compared to the phonons of the low symmetry phase. Note that the Z point of the primitive cell of the Pmmn phase is folded onto the Γ point of the supercell shown here. Thus, the antipolar mode in the primitive Z point is now at the Γ point slightly below the ferroelectric mode. The low symmetry phase shows no unstable modes.