Supplementary Materials for Observation of Double Weyl Phonons in the Parity Breaking FeSi

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I. INELASTIC X-RAY SCATTERING

Inelastic x-ray scattering directly measures the phonon dynamic structure factor, $S(\boldsymbol{Q},\omega)$, where $\boldsymbol{Q}$ and ω are the momentum transfer and phonon energy, respectively [1, 2]. This quantity is related to the imaginary part of the phonon susceptibility, $\chi''(\boldsymbol{Q},\omega)$, via the thermal Bose-factor as:

$$\chi''(\boldsymbol{Q},\omega) = S(\boldsymbol{Q},\omega)(1 - e^{-\omega/k_B T}). \quad (1)$$

All data that we shown in the main text has been converted from $S(\boldsymbol{Q},\omega)$ to $\chi''(\boldsymbol{Q},\omega)$ through Eq. 1.

II. FITTING OF PHONON SPECTRA

The data is fitted using a sum of the signals from different phonon modes and convoluted with the energy resolution function $R(\omega)$. Here $R(\omega)$ is found to be well described by a pseudo-Voight function

$$R(\omega) = (1-\alpha)\frac{A}{\sqrt{2\pi}\sigma}e^{-\frac{\omega^2}{2\sigma^2}} + \alpha\frac{A}{\pi}\frac{\gamma}{\omega^2+\gamma^2}. \quad (2)$$

We also include a constant offset I_c in our fitting routine, such that

$$\chi''(\boldsymbol{Q},\omega) = \sum_i \chi_i''(\boldsymbol{Q},\omega) * R(\omega) + I_c. \quad (3)$$

Here phonon modes are represented by the damped harmonic oscillator form [3]

$$\chi_i''(\boldsymbol{Q},\omega) = \frac{4A_i\gamma_i\omega\omega_i}{\pi\left[(\omega^2-\omega_i^2)^2 + 4\omega^2\gamma_i^2\right]}. \quad (4)$$

where A_i, ω_i and $2\gamma_i$ are the intensity, energy and full width at half maximum (FWHM) of phonon peak i at the measured momentum transfer, $\boldsymbol{Q}$. In all our fittings, the γ_i is found to be much smaller than the instrumental energy resolution, $\Delta E \sim 1.5$ meV, therefore all fitted peaks are essentially resolution limited.

Due to the finite instrumental energy resolution, peak separation smaller than 0.5 meV could not be experimentally resolved. Therefore the number of phonon peaks used in our fittings are based on the resolution-convoluted DFPT calculation.

III. SAMPLE GROWTH

Single crystals of FeSi were grown using a Ga flux method. The starting materials were mixed in a molar ratio of 1:1:20 in a glovebox filled with argon. The mixture was placed in an alumina crucible and sealed in a fully evacuated quartz tube. The crucible was heated to 1150°C and dwelled for 24 h, then cooled slowly to 850°C at 2 K/h, where the flux was centrifuged. The excess Ga flux on the faces was removed by the diluted hydrochloric acid.

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