Anharmonic Phonon Dispersion in Polyethylene

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Abstract: The second-order Green's function method for anharmonic crystals has been applied to an infinite, periodic chain of polyethylene taking into account up to quartic force constants. The frequency-independent approximation to the Dyson self-energy gives rise to numerous divergent resonances, which are fortuitous. Instead, solving the Dyson equation self-consistently with a frequency-dependent self-energy resists divergences from resonances or zero-frequency acoustic vibrations. The calculated anharmonic phonon dispersion, which nonetheless displays many true resonances, and anharmonic phonon density of states furnish hitherto unknown details that explain smaller features of observed vibrational spectra.