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Finding order in disorder: Raman spectroscopy of amorphous silicon, from ab initio to multiscale modeling[1]

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Amorphous silicon is an interesting material for photovoltaics, thermoelectrics, batteries, detectors, or thin-film transistors, whether as active material, a route to nano/microcrystalline silicon, or a passivation for crystalline silicon. Amorphous silicon is also a well-studied model system for amorphous materials in general, which present unique opportunities and challenges compared to crystalline materials. Raman spectroscopy is an important characterization tool for furthering our understanding and future applications. To enable its use for mapping local strain distributions, we calculated the Raman spectrum with ab initio density-functional perturbation theory under different strains and found peak shifts proportional to the trace of the strain. We showed this is the general form for isotropic amorphous vibrational modes, by symmetry analysis and explicit computation. These results were confirmed by accompanying experimental measurements. [1] Much larger structural models are needed for other interesting questions about a-Si:H, such as medium- and long-range order, nano/microcrystalline Si, amorphous/crystalline interfaces, or amorphous nanostructures. While quantum-mechanical calculations become infeasible, classical inter-atomic potentials can provide vibrations but need a model for Raman intensities. We retool the old idea of a bond-polarizability model by parametrization from our detailed database of ab initio Raman tensors, opening the way for large-scale Raman calculations on complex Si materials.

[1] David A. Strubbe, Eric C. Johlin, Timothy R. Kirkpatrick, Tonio Buonassisi, and Jeffrey C. Grossman, "Stress effects on the Raman spectrum of an amorphous material: theory and experiment on a-Si:H," Phys. Rev. B 92, 241202(R) (2015).

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