Spatial dispersion properties from DFPT: Dynamical quadrupoles and flexoelectric tensor

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In condensed-matter physics, spatial dispersion refers to the dependence of many material properties on the wavevector **q** at which they are probed, or equivalently on the gradients of the external field (electric, magnetic, strain...) and/or the response in real space. Remarkable examples of such gradient effects include the natural optical rotation, [1] whereby some crystals rotate the polarization plane of the transmitted light, or the flexoelectric tensor, [2] which describes the polarization response to a gradient of applied strain. The former is originated from the first-order wavevector dependence of the dielectric susceptibility tensor, while the latter can be written as the electrical current that is produced by an acoustic phonon at second-order in **q**. Density-functional perturbation theory (DFPT) appears as the ideal framework to compute these effects from first principles, but the general computational tools to deal with the long-wavelength limit are currently missing.

In this contribution we present two applications of a general formalism [3] based on an analytical long-wavelength expansion of the second-order DFPT energies that enables the direct calculation of spatial dispersion quantities. The latter are directly obtained from the uniform field linear-response functions and at essentially no additional cost. In particular, we elaborate on the specific implementation of the method for the cases of the flexoelectric tensor and the dynamical quadrupoles (the higher-order multi-polar counterpart of the Born effective charges). The flexoelectric coefficients calculated for several materials are directly benchmarked against previously published results, [4, 5] whereas the lattice sum of the PbTiO 3 quadrupole moments is shown to exactly reproduce its piezoelectric tensor, as predicted by R.M. Martin in his seminal paper. [6]

References

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