Theory and computation of electronic and nuclear response to applied electric fields in extended 1D-3D periodic systems

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The electronic and nuclear (structural/vibrational) response of 1D-3D nanoscale systems to electric fields gives rise to a host of optical, mechanical, spectral, etc. properties that are of high theoretical and applied interest. Due to the computational difficulty of treating such large systems it is convenient to model them as infinite and periodic (at least, in first approximation). The fundamental theoretical/computational problem in doing so is that the position operator normally employed for the interaction with the field breaks translational symmetry and, in addition, is unbounded. Several solutions have been suggested. Our own approach is to replace the usual scalar interaction potential with the time-dependent vector potential, which results in the so-called vector potential approach (VPA). The advantages of the VPA are that: (1) static and dynamic properties fall under the same rubric and (2) the formalism is particularly well-suited for adapting the algorithms of molecular quantum chemistry.

This talk will focus on the recent development and implementation of the VPA for a variety of applications. Among these applications are: (1) static/dynamic linear and nonlinear optical properties, including both electronic and vibrational contributions; (2) termination effects and surface charge; (3) piezoelectricity; (4) infrared and Raman vibrational intensities; and, tentatively, (5) combination with magnetic fields (optical rotation). Although implementation is feasible within plane wave as well as local basis set codes our efforts, thus far, have been confined to the latter in collaboration primarily with the CRYSTAL group in Torino. A large part of the computational methodology, in turn, has been developed in collaboration with Michael Springborg and co-workers in Saarbrucken.