Dielectric Response of Periodic Systems from Quantum Monte Carlo[1] PAOLO UMARI, DMSE-MIT, ANDREW J. WILLAMSON, GIULIA GALLI, Lawrence Livermore National Laboratory, NICOLA MARZARI, DMSE-MIT —

We introduce a novel approach to study the response of periodic systems to finite homogeneous electric fields using the diffusion Quantum Monte Carlo method. The interaction with the electric field is expressed through a generalized many-body electric-enthalpy functional; a Hermitian local potential is then constructed that determines the evolution towards the ground state. This local potential depends self-consistently on the Berry-phase polarization, and is evolved "on-the-fly" in the course of the simulation, with the polarization operator evaluated using forward-walking. To validate this approach we calculated the dielectric susceptibility of simple molecular chains, greatly over-estimated by standard density-functional approaches, and found good agreement with the results obtained with correlated quantum-chemistry calculations.

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