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**Linear and Non-Linear Dielectric Response of Periodic Systems from Quantum Monte Carlo**

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We present a novel approach that allows to calculate the dielectric response of periodic systems in the quantum Monte Carlo formalism. We employ a many-body generalization for the electric enthalpy functional, where the coupling with the field is expressed via the Berry-phase formulation for the macroscopic polarization. A self-consistent local Hamiltonian then determines the ground-state wavefunction, allowing for accurate diffusion quantum Monte Carlo calculations where the polarization's fixed point is estimated from the average on an iterative sequence. The polarization is sampled through forward-walking. This approach has been validated for the case of the polarizability of an isolated hydrogen atom, and then applied to a periodic system. We then calculate the linear susceptibility and second-order hyper-susceptibility of molecular-hydrogen chains with different bond-length alternations, and assess the quality of nodal surfaces derived from density-functional theory or from Hartree-Fock. The results found are in excellent agreement with the best estimates obtained from the extrapolation of quantum-chemistry calculations.

P. Umari, A.J. Williamson, G. Galli, and N. Marzari  
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