Theory of surface second-harmonic generation for semiconductors including effects of nonlocal operators

Sean M. Anderson, Nicolas Tancogne-Dejean, Bernardo S. Mendoza, Valerie Veniard.

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Abstract

We formulate a theoretical approach of surface second-harmonic generation from semiconductor surfaces based on the length gauge and the electron density operator. Within the independent particle approximation, the nonlinear second-order surface susceptibility tensor $\chi abc(-2\omega;\omega,\omega)$ is calculated, including in one unique formulation (i) the scissors correction, needed to have the correct value of the energy band gap, (ii) the contribution of the nonlocal part of the pseudopotentials, routinely used in ab initio band-structure calculations, and (iii) the derivation for the inclusion of the cut function, used to extract the surface response. The first two contributions are described by spatially nonlocal quantum-mechanical operators and are fully taken into account in the present formulation. As a test case of the approach, we calculate $\chi xxx(-2\omega;\omega,\omega)$ for the clean Si(001)2×1 reconstructed surface. The effects of the scissors correction and of the nonlocal part of the pseudopotentials are discussed in surface nonlinear optics. The scissors correction shifts the spectrum to higher energies though the shifting is not rigid and mixes the 1ω and 2ω resonances, and has a strong influence in the line shape. The effects of the nonlocal part of the pseudopotentials keeps the same line shape of $|\chi xxx2\times1(-2\omega;\omega,\omega)|$, but reduces its value by 15%–20%. Therefore the inclusion of the three aforementioned contributions is very important and makes our scheme unprecedented and opens the possibility to study surface second-harmonic generation with more versatility and providing more accurate results.