Ab initio approach to nonlinear optical response with strong excitonic effects: formulation and applications

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Nonlinear optical (NLO) responses play a key role in fundamental science and technological applications. The theory and calculations of NLO responses including electron-hole interactions (excitonic effect), which is crucial for low-dimensional materials, remain underdeveloped. In this talk, we present a novel ab initio approach for calculating NLO responses that incorporates excitonic effects using an exciton-state coupling and Berry phase framework. We employ a diagrammatic representation to elucidate the detailed nonlinear excitonic processes and the underlying selection rules. As applications of the new method, firstly, we demonstrate that the significant enhancement of second harmonic generation (SHG) in monolayer MoS2 and hBN is due to strong electron-hole interactions and specific excitons. In particular, our calculations reveal a prominent peak associated with C excitons in monolayer MoS2, in excellent agreement with recent experiments. Secondly, we propose a method to generate a pure spin DC current (with zero net charge current) in zigzag graphene nanoribbons (ZGNR), which are 1D antiferromagnets, via a second-order NLO process. Further, by applying an additional static electric field, we can energetically separate and selectively excite the up and down spin channels of the ZGNR, enabling fully spin-polarized photocurrent generation. These properties make the ZGNRs promising candidates as versatile spin sources for spintronics applications.

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