

Excitonic third harmonic generation in bilayer MoS₂ enhanced by static in-plane electric fields

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Abstract

We numerically and theoretically investigate the linear optical absorption and excitonic third harmonic generation (THG) for a bilayer MoS₂ in the presence of static in-plane electric fields. The linear absorption spectrum exhibits Stark shifts of the exciton transition energies as well as spectral broadening and a reduction of the height of the absorption peaks as a result of the hybridization of exciton states and exciton ionization induced by static in-plane fields. We solve the semiconductor Bloch equations including inter- and intraband excitations and excitonic many-body effects. Our results demonstrate that unexpectedly, the excitonic THG can be significantly enhanced by static fields as long as exciton ionization is not yet dominating. The amount of enhancement increases nonlinearly with the amplitude of the optical field. Within a perturbative analysis, we find that fourth-, fifth-, and higher-order contributions which represent transitions between bound and continuum states are involved in determining the excitonic THG. Without a static field, the third- and fifth-orders interfere destructively. With increasing strength of the static field this interference continuously becomes more constructive and consequently the excitonic THG is enhanced. This electric-field-induced enhancement might also be present for other nonlinearities and, furthermore, time-dependent electric fields could potentially be applicable to control the nonlinear dynamics.