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TDDFT approach to study nonlinear excitonic effects in Four-Wave Mixing processes¹ VOLODYMYR TURKOWSKI, Physics Department and NSTC, University of Central Florida, Orlando, FL 32816, MICHAEL N. LEUENBERGER, Physics Department and NSTC, University of Central Florida, Orlando FL 32816 — We develop a time-dependent density-functional theory (TDDFT) formalism to study nonlinear effects in the processes in Four-Wave Mixing experiments. Namely, we generalize our recently proposed approach to calculate excitonic and biexcitonic eigen-energies within TDDFT on the dynamical case, which includes nonlinear effects, the like exciton-exciton interaction. In particular, we derive the TDDFT version of the nonlinear time-dependent equation for excitonic polarization with terms which correspond to exciton-exciton correlations through the memory functions. We have obtained the formula which relates the memory functions with the TDDFT exchange- correlation (XC) kernel. To test the formalism, we calculate the 2D Fourier spectra of a GaAs multi-quantum well system and compare them with experimental results in the case of several XC kernels. In addition, we compare the results with the ones obtained within a many-body method for non-linear effects in semiconductors. It is shown that the results obtained within the TDDFT approach may reproduce semi-quantitatively the 2D Fourier spectra, including the nonlinear effects, in the case of several phenomenological potentials.

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