Time-dependent Density Functional theory in the nonlinear domain: successes and failures

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The time-dependent density-functional theory (TDDFT) has been documented as a "pretty good" computational approach for the linear response to external fields, but its utility in the nonlinear domain has not been studied as thoroughly. My talk will consider several nonlinear phenomena and discuss how well the TDDFT performs. The simplest nonlinear observables are hyperpolarizability and nonlinear susceptibility, and here the TDDFT is found to be quite useful. The second area is the interaction of strong laser pulses on media, including coherent phonon generation and field-dependent diectric functions. Here again the TDDFT offers an attractive compromise between computational tractability and predictive power. For some other processes, attempts to use TDDFT have little or mixed success. Namely, attempts to describe Rabi oscillations in the TDDFT framework have been unsuccessful, while attempts to treat the effects of electron relaxation in X-ray absorption spectra (XAS) are mixed. While a TDDFT approach to Langreth's cumulant treatment has been successfully employed to describe inelastic losses, excitonic effects are beyond the reach of TDDFT.