After revisiting the theory of pump-probe photoabsorption spectroscopy we propose a nonequilibrium Green’s function (NEGF) approach to calculate the transient spectrum of nanoscale systems. We can deal with arbitrary shape, intensity, duration and relative delay of the pump and probe fields and we can include ionization processes as well as hybridization effects due to surfaces. We present numerical simulations of atomic systems using different approximate self-energies and, whenever possible or available, find good agreement with CI calculations and experiments. The NEGF approach offers a first-principle methodology to predict and interpret pump-probe photoabsorption spectra of systems that are out of reach with other methods. In the last part of the talk we will discuss future challenges and reachable goals.