Nonequilibrium optical response of metals irradiated by ultrafast laser pulses

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Ultrafast light coupling with metal surfaces shows strong potential for nanostructuring applications relying on the capacity to localize light energy on the nanoscale. Controlling light confinement requires to understand the transient variation of the optical response during ultrafast irradiation. The fundamental approach we propose based on ab initio calculations.allows elucidating the influence of carrier nonequilibrium on optical properties. The challenges are both to evaluate the optical response under strong electron-phonon nonequilibrium and to investigate the primary processes responsible for the optical change during laser-solid interaction. Calculations are carried out in the framework of the density functional theory associated to quantum molecular dynamics. Our results shed light on the intricate role of electronic structure modifications and possible optical transitions, driving the laser energy absorption into the material. The revealed key processes based on Fermi smearing on an evolving density of states are of paramount interest for controlling laser energy deposition, surface plasmon excitation and subsequent surface nanostructuring. The calculations predict the possibility of an ultrafast laser-driven plasmonic switch on a typically non-plasmonic material (W), confirmed by pump-probe ellipsometric measurements [1]. The consequence of our results is far reaching as they propose also a route for achieving the highest energy confinement under ultrashort laser irradiation.

[1] E. Bévillon, J.P. Colombier, V. Recoules, H. Zhang, C. Li, R. Stoian, "Ultrafast switching of surface plasmonic conditions in nonplasmonic metals", Physical Review B 93 (16), 165416 (2016).