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Non-Equilibrium theory of hot electron generation in plasmonic nanostructures under illumination – thermal vs. non-thermal effects

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Abstract

Understanding the interplay between electrons, photons and phonons is a fundamental problem in physical chemistry. Recently, interest in this problem resurfaced in the context of non-equilibrium (hot) electron distributions, which are key for applications such as photo-catalysis, sensing, up-conversion etc.. Here, we report a formulation of the theory of hot electron generation in plasmonic nanostructures under continuous wave illumination, taking into account non-equilibrium as well as thermal effects. Specifically, we consider the effect of both photons *and* phonons on the electron distribution function, and calculate self-consistently the increase in electron and lattice temperatures above ambient conditions (as observed experimentally). This enables us to go well beyond the limits of existing theories, which are limited to low illumination intensities. We determine the electronic distribution and deviations from equilibrium under different conditions, and evaluate the rise in electron and lattice temperatures. Doing so, we correct the multitude of errors in existing formulations of the problem. Finally, we discuss the prospect of using the hot electrons for photocatalysis in light of recent experiments, and identify the efficiency and the photocatalytic performance.

Date & Location:

Tuesday, December 4, 2018, 11:00
Lecture room, Physics Building (ground floor)