Photoinduced Electron Dynamics in Nanostructures: Nonuniform and Self-Consistent Light-Matter Interactions

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In the conventional optical responses of molecules, wavelengths of the lights are usually considered to be much longer than molecular sizes. Thus, a target molecule is well approximated by a point dipole and the dipole feels an almost uniform electromagnetic field. This condition underlies the conventional dipole approximation. Furthermore, light is an external field to excite molecules and its wavelength is definitely determined by an apparatus condition. Since spatial resolution of spectroscopy is limited by the wavelength of the incident light, it is impossible to gain molecular properties in a local region shorter than the wavelength, i.e., diffraction limit. However, recent development of nanofabrication and nano-optical techniques requires a more general optical response theory fully taking account of nonuniform and self-consistent light-matter interactions.

First, a generalized theoretical description of a light-matter interaction beyond a dipole approximation is developed on the basis of the multipolar Hamiltonian with the aim of understanding the near-field excitation in nanostructures[1]. The theory is formulated for a system consisting of a molecule and a near-field, where a nonuniform electric field plays a crucial role. A nonuniform electronic excitation of a molecule is demonstrated by solving a time-dependent Kohn-Sham equation in real-space and real-time [2,3] with an implementation of the nonuniform light-matter interaction.

Second, optical forces induced by a near-field are calculated for a 1 nm-sized metal particle mimicked by a jellium model fully taking account of multipole interaction [4]. A highly localized near-field nonuniformly polarizes the metal particle. The locally induced polarization charges in the molecules are partly canceled by the screening charges. The polarization and screening charges generally contribute to the attractive and repulsive forces, respectively, and a sensible balance between these charges results in several peaks in the optical force as a function of the frequency of the near-field. The resonance excitation does not necessarily maximally induce the net force and the force exerted on the molecules strongly depends on the details of their electronic structures.

Finally, I will discuss our recent research activity of electron and electromagnetic field coupled dynamics in nanostructures.

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