Probing Electron Dynamics with Ultrafast X-Ray Scattering

Content

In the past decade, the advent of x-ray free-electron lasers has enabled ultrafast non-resonant x-ray scattering experiments to track photochemical reactions in real time [1] and to measure the change of the molecular electronic density upon photoexcitation [2]. These experiments offer information complementary to spectroscopic techniques with particular sensitivity to molecular structure. The recent increase in repetition rate of facilitites such as the European XFEL and LCLS-II and the anticipated further decrease in pulse duration will make it possible not only to track the nuclear but also the electronic motion on the femto- and eventually attosecond time scale. [3,4] Here, we will present simulations of ultrafast x-ray scattering from a molecular electronic wave packet, demonstrate that the signal can probe electronic coherence, and discuss the underlying theory. [5–7] We will explain how these signals can be calculated from ab-initio electronic structure theory and quantum dynamics simulations. We will furthermore address important aspects of experiments such as rotational averaging and energy-integrating detectors that have to be considered to reliably predict the observable signal and to guide the prospective first observation of electron dynamics with ultrafast x-ray scattering.

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Contribution Type: Hot topic contribution