Ultrafast structural changes in matter induced by intense X-ray free-electron laser pulses

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X-rays have become established as an invaluable probe for gaining an atomic insight into the structure of matter through various kinds of interaction processes, such as scattering, absorption, and emission of photoelectrons and fluorescence. Since these interactions were usually weak with the previous X-ray sources, X-ray irradiation was assumed not to modify matter. This situation has been altered by the recent advent of X-ray free-electron lasers (XFELs), which can generate brilliant femtosecond X-ray pulses.

When an XFEL pulse irradiates matter, photoelectrons and Auger electrons are emitted during or shortly after the irradiation of the pulse and trigger cascades of secondary electrons. If the radiation dose exceeds a critical value, the electron excitations strongly change interatomic potential surface and cause subsequent atomic disordering and may even lead to the Coulomb explosion in the case of high X-ray dose. Given the time scale of electron cascading (typically, a few tens of fs) and inertia of atoms, the onset of atomic disordering is expected to take place behind the start of X-ray exposure. Indeed, it has been predicted that ultrafast X-ray pulse as short as ~10 fs with sufficient intensity can produce high-quality diffraction before the onset of substantial radiation damage, enabling structure determination of macromolecular nanocrystals and even individual biomolecule [1]. A deep understanding of transient XFEL interaction with matter is essential not only because of fundamental interest but for analyzing experiments with intense XFEL pulses.

Up to now, transient XFEL-matter interactions have been relying on theoretical modeling, validated by time-integrated measurements of charge states of ions and emitted fluorescence using a single XFEL pulse. To observe time-dependent X-ray interactions with matter, we developed a femtosecond X-ray pump-X-ray probe method [2] by combining nano-focusing optics [3] and twin XFEL pulses with controlled time separations [4] at SPring-8 Angstrom Compact free-electron LAser (SACLA) [5]. This method was applied to various materials (diamond [2,5], silicon [6], oxides, and protein crystals) and revealed the time scale of the electron excitations and the onset time of the structural changes.

In this talk, the XFEL-induced transient structural changes in matter revealed by the pump-probe experiments are discussed. In addition, preliminary results of the advanced pump-probe experiments using seeded-XFEL pulses [7,8] will be presented.

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