X-ray Two-Photon Absorption Spectroscopy

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The advent of X-ray free-electron laser makes it possible to observe various multi-photon absorption processes even in the hard X-ray region [1-7]. Among them, two-photon absorption (TPA) is the lowest order and, hence, is the most efficient and easy-to-use. Theoretically, it is categorized into two types: direct TPA and sequential TPA.

First, we discuss the direct TPA, where the intermediate state between two absorption processes is virtual. The selection rule of the direct TPA is different from that for one-photon absorption (OPA) used in the conventional X-ray absorption spectroscopy (XAS) [3]. X-ray spectroscopy with TPA is expected to reveal hidden electronic state from the conventional XAS. However, the TPA process has been observed using quite intense X-rays at the cost of severe sample damage [3,8], and thus, cannot probe the undamaged state.

In this paper, we show that TPA can be measured with much weaker beam, and report the first X-ray TPA spectrum for metallic copper. We also study the damage threshold for XAS, and conclude that the measured TPA spectrum is almost free of damage.

Next, we discuss the sequential TPA, where the intermediate state is real. In this case, the first photo-ionization may open a new absorption channel for the second X-ray photon, which cannot be accessed in the ground state [7,9,10]. Using a sequential process, we report the resonant absorption spectrum at the 1s-2p transition of copper for the first time. We discuss the difference between the 1s-2p resonant absorption spectrum and the 2p-1s fluorescence spectrum.

References

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