Occupied density of states from single shot two-color measurements

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To experimentally determine the occupied electronic structure of materials with elemental selectivity, one often employs RIXS or X-ray emission spectroscopy. Especially in the soft X-ray range, these methods are limited in their efficiency. To get one photon on the detector, typically 10^8 X-ray photons have to impinge on the sample. Here, the low fluorescence yield of 0.1-1% in the soft X-ray range and the low angular acceptance of grazing incidence grating spectrometers poses the biggest limits.

With high intensity pulses from free-electron lasers (FELs), one can overcome these issues for example by using stimulated emission processes [1-3]. With a higher photon energy, core excitations are created and with a lower photon energy a stimulation cascade is triggered. Seeded free-electron lasers deliver pulses of well-controlled spectral content, even of two different colors in a single pulse. The spectroscopic information can straightforwardly be derived from the transmitted intensity.

With soft X-rays on solids, Auger decays dominate the core decays and create fast electrons in the sample that in turn create a multitude of valence excitations. On a timescale shorter than the average core-hole lifetime, holes appear in the valence band with a distribution analogous to the density of states [4]. These holes can be efficiently probed via X-ray absorption spectroscopy and yield the required information on the usually occupied density of states of a material.

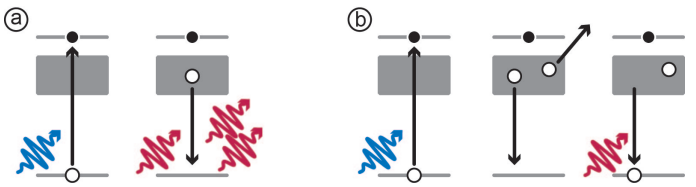

Figure 1: (a) Stimulated X-ray emission amplifies the probing redshifted beam. (b) On a solid, Auger processes create holes in the valence states that are probed with the redshifted beam.

References

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