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Resonance-enhanced XUV-NIR four-wave mixing

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While wave-mixing processes are widely exploited in the IR and optical regime, attempts to mix IR/optical photons and XUV or soft X-ray photons have hitherto been hampered.
A spectroscopy based on sum- and/or difference-frequency generation (SFG/DFG) around soft X-ray or XUV resonances promises, like Resonant Inelastic X-ray Scattering (RIXS), detailed spectroscopic insights on low-energy excitations within the material of interest, while overcoming the critically low signal levels that constitute the core limitation of RIXS.
We show preliminary results of an experiment at the free-electron laser in Hamburg (FLASH) on lithium fluoride (LiF) observing both SFG and DFG involving one XUV photon around the Li+ 1s-2p resonance (62 eV) and two 800 nm NIR photons in reflection from a LiF single crystal ($\omega = \omega_{XUV} \pm 2\omega_{IR}$). Within the experimental sensitivity, the corresponding processes involving the mixing of only one optical photon with an XUV photon ($\omega = \omega_{XUV} \pm \omega_{IR}$) were not observed.
We observe SFG and DFG during temporal overlap of the XUV and IR laser pulses as well as a strong dependence on the XUV photon energy explainable through the presence of the core level excitonic resonance. To develop an understanding of the wave-mixing process involving NIR and XUV photons, we discuss the role of material resonances and phase matching conditions as well as upcoming experimental efforts to explore them further.

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