Mixing x-ray and optical photons to directly probe valence charges

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Abstract: We propose wavemixing of x-ray and optical photons as a nonlinear method to probe valence charges. Specifically, we report on theoretical and experimental approaches to access nonlinear frequency conversion from x-rays into lower energy photons.

Wavemixing and frequency conversion processes are well known in the optical regime and find their applications throughout a broad field of research and technology. Extending these effects into the x-ray domain remains challenging however, mainly due to their low conversion efficiencies. Yet, with the ascent of new generation synchrotrons and x-ray free electron lasers (FELs), these processes become progressively accessible.

In the presented work, we focus specifically on the conversion of x-rays into lower energy photons (XUV to optical). For example, in the process of x-ray parametric down-conversion, an x-ray photon (10 keV) can be converted into an x-ray signal photon at slightly lower energy (10 keV – ΔE) and an idler photon at the corresponding energy difference (ΔE ~ 5 – 200 eV). Intriguingly, this nonlinear conversion effect couples predominantly to the valence electrons of the irradiated matter, as Freund noted almost 50 years ago [1]. Exploring the effect on a first-principles basis, we have developed a theoretical framework based on nonrelativistic Quantum electrodynamics. Using this, we can identify the nonlinear material response that mediates the conversion and connect it to experimental observables. Our simulations (for crystalline materials) indicate that the response correlates with the localization of the valence electrons (Figure 1 right).

This selective coupling can be used to image the valence charges with atomic scale resolution – similar to conventional x-ray diffraction.

In addition, this diffractive method is spectroscopically sensitive: By selecting the idler energy ΔE accordingly, electronic transitions of the valence charges may be probed. Ultimately, this could provide access to transition charge densities at atomic scale resolution inside the bulk material.

We present an overview of our current theoretical [2] and experimental work [3,4] to explore this conversion of x-rays into lower energy photons. We illustrate open challenges and potential future applications for nonlinear frequency conversion as a novel probing method.

References

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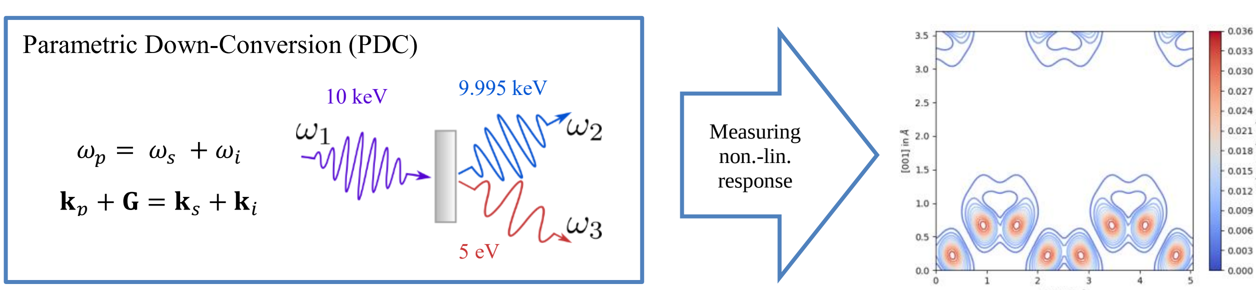

Figure 1 : Parametric conversion of x-rays into signal (ω_2) and idler (ω_3) photons (left). The phase-matching condition is given by energy and momentum conservation. The nonlinear response function for diamond (right) exhibits a strong response where the probability density of valence electrons is high.