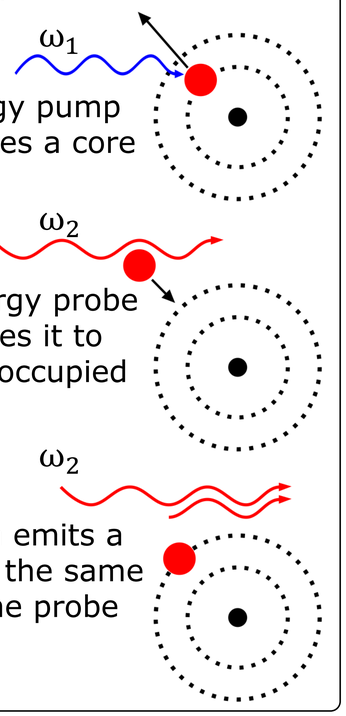
Two Color Stimulated X-ray Raman Scattering

SLAC NATIONAL ACCELERATOR LABORATORY

Bryce Eggers, Andy Aquilla

Introduction

Stimulated X-ray Raman Scattering (SXRS) is an inelastic two-photon process.



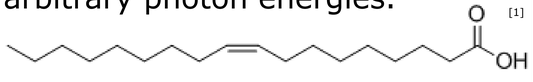
A high energy pump photon ionizes a core electron

A lower energy probe photon causes it to fall to an unoccupied level

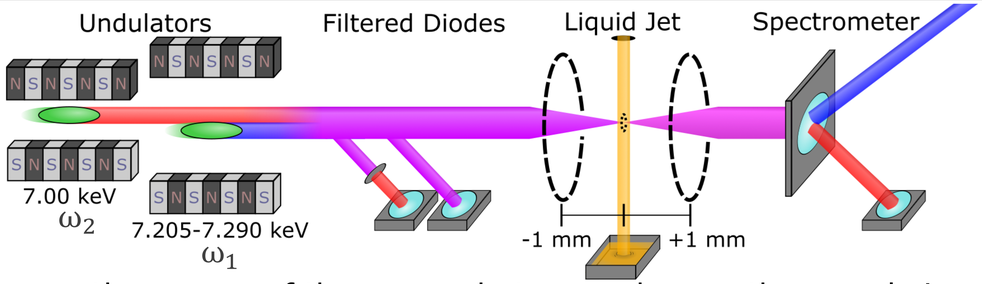
The electron emits a photon with the same energy as the probe

Previous studies on SXRS have been limited by the inability to supply both pump and probe photons simultaneously.

New improvements at LCLS allow us to create a beam tuned to two arbitrary photon energies.

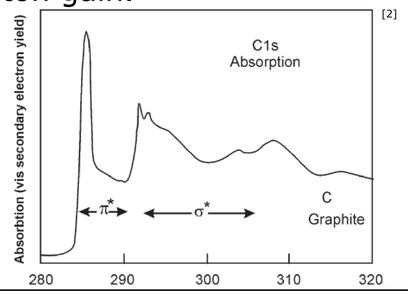


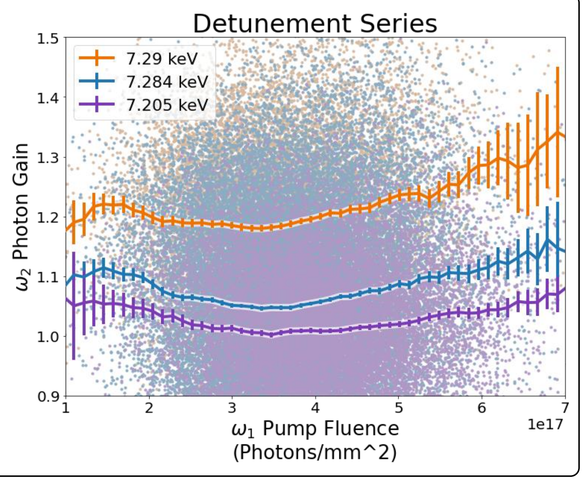
We sought to demonstrate SXRS using oleic acid with a carbon k-edge of around 290 eV.



We can vary the energy of the pump photons and move the sample in and out of focus. To detect SXRS, we measured the number of pump and probe photons before scattering and the number of probe photons after, allowing us to calculate the gain in probe photons.

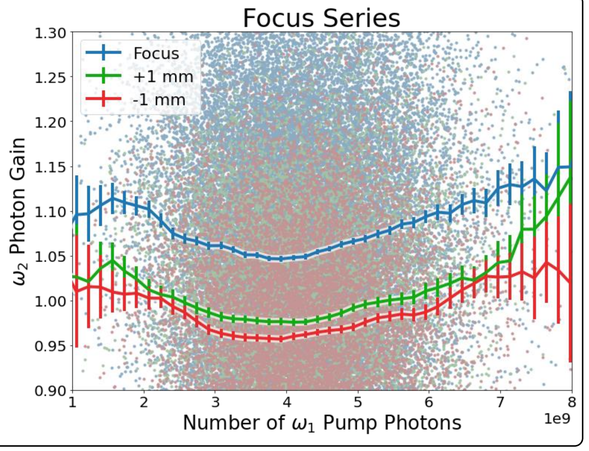
As the difference between the pump and probe photon energy approaches the expected k-edge, we see a 6% and 19% increase in photon gain.

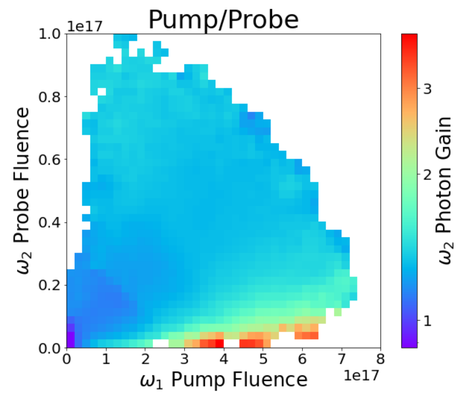




Since SXRS is a nonlinear two-photon process, it not only depends on the number of photons that hit the sample but also on the number of photons that hit approximately the same spot.

When the sample moves into focus, increasing the fluence with the same number of photons, we see an 8% increase in photon gain.





There is a distinct gain region, although competing processes and saturation effects may obscure the expected bilinear relationship.

Conclusions

We can clearly see a non-linear effect tuned to the expected k-edge of oleic acid, strongly indicating the existence of SXRS.

Since stimulated XRS is much easier to detect than its spontaneous counterpart, this method will drastically speed up Raman spectroscopy, allowing for the study of systems that change over short periods of time.

Acknowledgments

Special thanks to Franklin Fuller, Brandon Hayes, Roberto Alonso-Mori, Alberto Lutman, Uwe Bergmann, and Dimosthenis Sokaras for their collaboration on the project.

Figure credits:
[1] ChemSrc
[2] Grant S. Henderson, et al.; X-ray Absorption Near-Edge Structure (XANES) Spectroscopy.