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Time-resolved second harmonic generation on Cu2O Rydberg excitons

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For the last decade, highly excited (Rydberg) states of electron-hole bound states (excitons), especially in in cuprous oxide (Cu₂O) [1], have been showing more and more similarities with their atomic relatives, enabling the exploration of Rydberg physics in a semiconductor setting. This talk will describe recent experimental results where a second harmonic generation (SHG) process, made resonant with Rydberg states of Cu2O excitons [2], is time-resolved with subpicosecond resolution.

This technique not only enables the direct measurement of the Rydberg states lifetimes but also reveals several intriguing dynamics, including the existence of two different mechanisms for SHG, an accelerated decay for large exciton density and traces of coherence lasting up to the state lifetime. Directly measuring the lifetime also enables to check for inhomogeneous broadening in the current high-precision spectroscopic data [3], which participates in charting the possibilities offered by Cu2O for future solid-state Rydberg technologies.

- [1] Kazimierczuk, T., Fröhlich, D., Scheel, S., Stolz, H. and Bayer, M., 2014. Giant Rydberg excitons in the copper oxide Cu2O. Nature **514**(7522), pp.343-347.
- [2] Mund, J., Fröhlich, D., Yakovlev, D. R., & Bayer, M. (2018). High-resolution second harmonic generation spectroscopy with femtosecond laser pulses on excitons in Cu2O. Physical Review B **98**(8), 085203.
- [3] Rogers, J.P., Gallagher, L.A., Pizzey, D., Pritchett, J.D., Adams, C.S., Jones, M.P., Hodges, C., Langbein, W. and Lynch, S.A., 2022. High-resolution nanosecond spectroscopy of even-parity Rydberg excitons in Cu 2 O. Physical Review B **105**(11), p.115206.