LABORATOIRE INTERACTIONS, DYNAMIQUES et LASERS
EMR9000 CEA, CNRS, Université Paris-Saclay

SEMINAIRE LIDYL

Hugo Marroux
LIDYL / ATTO

Le Jeudi 29 Septembre 2022 à 11h
Orme des Merisiers, Bât.701, Pièce 17 (salle de séminaires)

The seminar will be accessible online on Zoom at the following address:
https://cnrs.zoom.us/j/97095984730?pwd=UU4yWXRad252NUF2OUlDWWZvTmQrUT09
ID de réunion : 970 9598 4730 / Code secret : T4ZHNg

Attosecond spectroscopy of core-excited molecules

Excitation of molecules with high-energy radiation removes electrons located in the inner electronic shell of the system. The following metastable state decay is an electronic process that occur in only a few femtoseconds and has so far only been studied in the frequency domain. I will present how we can use attosecond transient absorption spectroscopy (ATAS) to first study and then manipulate these states in the time domain. ATAS is principally employed to observe light matter interaction at relatively high laser intensities.[1,2] By tuning the intensity we show that core-excited state (CES) lifetimes can be measured in atomic[3] and molecular systems.[4] This time domain measurement reveals hidden dependences of the CES lifetimes on the core orbital alignment in the molecular frame. This dependence is a clear evidence of interatomic electron transfer mechanisms.

I will then show how ATAS can be employed to observe sub-natural linewidth information in atomic and molecular systems. By implementing background subtraction and noise reduction routines, the measurement sensitivity was brought close to the detector's noise floor and quasi-removed laser induced noise. This improvement allowed us to observe small absorption changes and revealed a time-dependent narrowing of absorption bands below their natural broadening limit. By comparing the narrowing observed in CH3I, ICl and atomic iodine at the $N_{4,5}$ edge, the so-revealed line splitting can be attributed to degeneracy lifting of the core levels under the ligand field interaction. This lineshape narrowing is explained in terms of perturbed free induction decay of the system's response and is a general feature of attosecond transient absorption. In the future, this new approach could be applied to other elemental edges but also to solid and liquid phase targets.

1. Ott, C. *et al.* Lorentz Meets Fano in Spectral Line Shapes: A Universal Phase and Its Laser Control. *Science (80-.).* **340**, 716–720 (2013).
2. Reduzzi, M. *et al.* Polarization control of absorption of virtual dressed states in helium. *Phys. Rev. A* **92**, 33408 (2015).
3. Bernhardt, B. *et al.* High-spectral-resolution attosecond absorption spectroscopy of autoionization in xenon. *Phys. Rev. A - At. Mol. Opt. Phys.* **89**, 23408 (2014).
4. Marroux, H. J. B. *et al.* Attosecond spectroscopy reveals alignment dependent core-hole dynamics in the ICl molecule. *Nat. Commun.* **11**, 5810 (2020).