Magnetic-only optical spectroscopy with ultrafast laser pulses

Lorenz Grünewald[1,2], Rodrigo Martín-Hernández[3], Elizaveta Gangrskaia[4], Alessandra Bellissimo[4], Carlos Hernández-García[4], Sebastian Mai[1]

[1] *Institute for Theoretical Chemistry, Faculty of Chemistry, University of Vienna, Austria*

[2] *Vienna Doctoral School in Chemistry (DoSChem), University of Vienna, Austria*

[3] *Grupo de Investigación en Aplicaciones del Láser y Fotónica (ALF-USAL), Dpt. Fisica Aplicada, Universidad de Salamanca, Spain*

[4] *Institute for Photonics, Faculty of Electrical Engineering and Information Technology, TU Wien, Austria*

lorenz.gruenewald@univie.ac.at

In optical spectroscopy, the vast part of light-matter interaction is constituted by the electric field (EF) of light interacting with the molecular electric dipole moment, as the analogous magnetic field (MF) interaction is orders of magnitude weaker. Still, magnetic dipole transitions can provide valuable complementary information due to their distinct spectroscopic selection rules. Nowadays, tightly focussed azimuthally polarized beams (APBs) can locally enhance the MF while suppressing the EF, resulting in a high MF/EF contrast along the beam propagation axis [2]. Previously reported simulations [1] revealed that upon focusing ultrashort APB pulses on a small metal iris, a so-called aperture, few-femtosecond ring currents are induced inside the aperture material, which in turn enhance the MF on the beam axis.

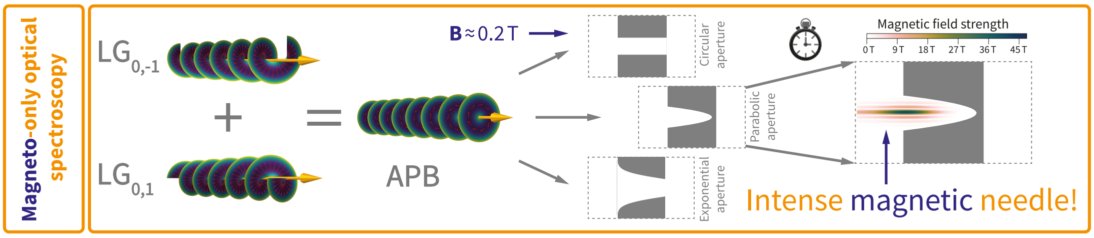

Figure 1: Illustration of MF enhancement within a magneto-only optical excitation scheme. In the simulations, the APB is constructed as a superposition of two Laguerre-Gauss (LG) beams which is focused onto a metallic aperture. Simulations for a set of different geometrical aperture shapes show high MF enhancement (heatmaps of **B** amplitude) and EF suppression (not shown).

In this contribution, we present particle-in-cell simulations of the spatiotemporal evolution of few-femtosecond APB pulses interacting with a conducting metal aperture. We investigated different aperture profiles with the objective of achieving a high MF enhancement and providing a large area of substantial MF/EF contrast. Moreover, we considered smooth aperture profiles that can be produced with nanofabrication techniques. For optimized aperture geometries, our findings predict MF strengths close to the beam axis that surpass the unapertured case by a factor of up to 70 at vanishing EF strengths. Furthermore, we provide evidence that neither the variation of the width of the APB pulse nor the variation of its wavelength within the visible regime leads to significant changes in the MF enhancement or in EF suppression. Our results indicate that a conceptually simple apertured APB setup can be utilized to investigate a wide range of atoms or molecules by means of a magneto-only optical spectroscopy.

References

[1] M. Blanco et al., Ultraintense Femtosecond Magnetic Nanoprobes Induced by Azimuthally Polarized Laser Beams. ACS Photonics, 6(1), (2019), 38–42

[2] M. Veysi, C. Guclu, and F. Capolino, Vortex beams with strong longitudinally polarized magnetic field and their generation by using metasurfaces. J. Opt. Soc. Am. B, 32(2), (2015), 345