## Ultrafast nonlinear effects in organic compounds and organic hybrid materials

H. Gonçalves<sup>1</sup>, Luís Alves<sup>1</sup>, Etelvina de Matos Gomes<sup>1</sup>, Rosa Cristina Moutinho Ferreira<sup>2</sup>, Susana Costa<sup>2</sup>, Manuela Raposo<sup>2</sup>, and Michael Besley<sup>1</sup>

<sup>1</sup> Center of Physics, University of Minho, Campus de Gualtar, 4710-057 Braga, Portugal <sup>2</sup>Center of Chemistry, University of Minho, Campus de Gualtar, 4710-057 Braga, Portugal goncalves.hmc@ua.pt

Synthesis and solution-processing of specialized chemical compounds can facilitate the fabrication of nano-electronic devices. Many of these emerging applications require organic materials with wellcharacterized optical properties, specifically, large two-photon absorption (2PA) and nonlinear optical (NLO) responses. The most common strategy to achieve a strong second order nonlinear optical response is to link donor and acceptor moieties on opposite ends of a  $\pi$ -conjugated aromatic spacer. This configuration promotes spatial charge transfer and a strong difference in dipole moments between the ground and excited electronic states. Individual organic molecules, tailor made following this strategy, can often exhibit exceptionally large intrinsic nonlinear optical properties, but the translation to a macroscopic electro-optic activity is often extremely difficult. Roughly more than 80% of all  $\pi$ -conjugated organic molecules crystallize in centrosymmetric space groups producing materials with no second order bulk susceptibility. Our group has demonstrated that is possible overcome this restriction by embedding organic molecules, which normally crystallize in centrosymmetric structures, into nanostructured polymeric matrix[1]. Recently, we observed greatly enhance the nonlinearities of nominally weak nonlinear materials when insert in a polymeric nanofibers, one-dimensional nanostructures (See figure 1). These types of nanostructures are attracting attention because of the unique optical interactions that arise from their sub wavelength size[2].

To explore this phenomena in detail we take advantage of our state of the art custom built Fluorescence Lifetime Imaging Microscope (FLIM) [3]. The temporal resolution of the FLIM is about 25 picoseconds and less than a 1 micron of spatial resolution. This system allows us to monitor the effects of nano-structured environments and their localized optical response. Other systems of current interest in our research group are graphene quantum dots both in solution and dispersed in a thin film of PVA, the interaction of excited dye molecules with a single layer graphene surface and tailor designed molecules for two photon absorption in the near infrared portion of the spectrum.

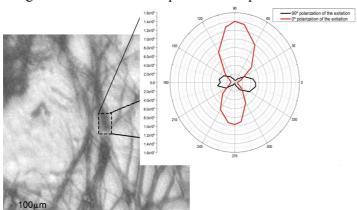


Figure 1 - Polarized light of the second harmonic generation of a nanofiber of poly(L-lactic acid) (PLLA) doped with para-nitroaniline. The red curve correspond to excitation using a polarization parallel to the fibres, the black curve correspond an orthogonally polarization excitation.

[1] D. V. Isakov, M.S. Belsley, E. de Matos Gomes, H. Gonçalves, P. Schellenberg, B.G. Almeida, Intense optical second harmonic generation from centrosymmetric nanocrystalline para-nitroaniline, Appl. Phys. Lett. 104, 181903-181907 (2014).
[2] J. Brewer, M. Schiek, I. Wallmann, H.G. Rubahn, First order optical nonlinearities χ<sup>2</sup> for organic manofibers from functionalized para-

pheny lenes Opt. Commun. 281, 3892-3896 (2008).

[3] C. Bernardo, et al., A versatile fluorescence lifetime imaging system for scanning large areas with high time and spatial resolution, in: M.F.P.C. Martins Costa, R.N. Nogueira (Eds.), Second Int. Conf. Appl. Opt. Photonics, International Society for Optics and Photonics, 928637-928641, 2014.