## Combination of Electron Energy-loss Spectroscopy and Orbital Angular Momentum Spectroscopy. Applications to Electron Magnetic Chiral Dichroism, Plasmon-loss, and Core-loss

Giovanni Bertoni<sup>1</sup>, Enzo Rotunno<sup>1</sup>, Amir Tavabi<sup>2</sup>, Matteo Zanfrognini<sup>3</sup>, Paolo Rosi<sup>3</sup>, Stefano Frabboni<sup>3</sup>, Ebrahim Karimi<sup>4</sup>, Rafal Dunin-Borkowski<sup>5</sup> and Vincenzo Grillo<sup>1</sup>

<sup>1</sup>CNR - Istituto Nanoscienze, Modena, Emilia-Romagna, Italy, <sup>2</sup>Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons, Jülich, Nordrhein-Westfalen, Germany, <sup>3</sup>Università di Modena e Reggio Emilia, Modena, Emilia-Romagna, Italy, <sup>4</sup>University of Ottawa, Ottawa, Ontario, Canada, <sup>5</sup>Forschungszentrum Juelich, Juelich, Nordrhein-Westfalen, Germany

Electron energy-loss spectroscopy (EELS) is a powerful technique for measuring the energy state of electrons after scattering. It has had incredible success in measuring the atomic and chemical properties of materials, allowing chemical maps to be recorded with atomic spatial resolution. Energy information alone is often sufficient to obtain information about chemical species, plasmons or even phonon excitations, especially when using high-energy-resolution spectrometers and gun monochromators. However, it is sometimes desirable to add a second dispersion variable, either in the form of momentum and energy double-dispersion EELS, or by acquiring a spectrum for a given momentum transfer.

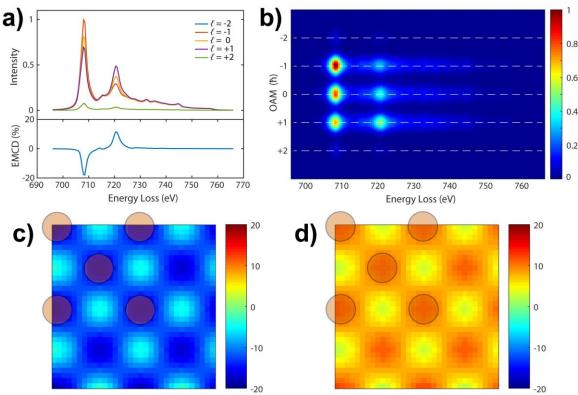
Here, we show that an orbital angular momentum (OAM) sorter [1] can be used to achieve simultaneous energy and OAM measurements. The motivation for the development of such a double-dispersion device is electron magnetic circular dichroism (EMCD). [2] Although such a technique is cumbersome for linear momentum (q, E) dispersed EELS, it is natural in the OAM basis  $(\ell, E)$ , in which magnetism introduces an asymmetry between the  $\ell = -1$  and  $\ell = +1$  channels.

Figure 1 shows a predicted combined OAM-EELS ( $\ell$ , E) spectrum for the L<sub>2,3</sub> core ionization edge of iron. According to our simulations, it should be possible to produce atomic-resolution EMCD maps. [3] This result would be of immense practical interest. The channeling should permit analysis of the results in terms of electronic properties to be simplified.

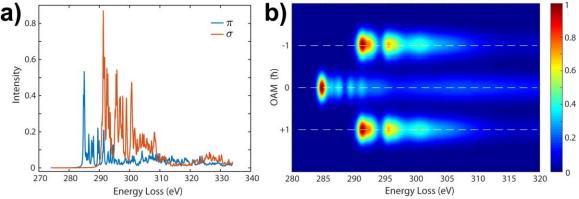
Beyond EMCD, we believe that there are prospects to use the same idea for the analysis of plasmon resonances [4] and core transitions. Figure 2 shows an advantage of applying OAM sorter to 2D anisotropic materials (h-BN, graphene, ...). According to simulations based on the symmetry of the projected density of states (pDOS), as implemented in standard density functional theory methods, combined OAM-EELS spectra should show a distinct difference between  $1s \to \sigma$  ( $\ell = -1, +1$ ) and  $1s \to \pi$  ( $\ell = 0$ ) transitions for the C-K edge. The advantage of the sorter would be to allow for direct separation of the two contributions in a single acquisition, whereas normally this separation would require tilting of the specimen in the standard (q, E) basis to separate the two overlapping features, which may be not possible experimentally.

This work is supported by Q-SORT, a project funded by the European Union's Horizon 2020 Research and Innovation Program under grant agreement No. 766970.





**Figure 1.** (a) Simulation of EMCD Fe-L2,3 edge spectra in Fe (bcc). (b) Double dispersion OAM-EELS in STEM mode for an [001]-oriented Fe (bcc) crystal). The EMCD signal is proportional to the difference between the  $\ell = +1$  and  $\ell = -1$  spectra. (c, d) Simulated STEM maps with predicted atomic spatial resolution obtained from the L3 and L2 EMCD signals, respectively (%).



**Figure 2.** (a) Calculated  $\pi$  and  $\sigma$  components for the C-K edge spectrum in graphene. (b) Double dispersion OAM-EELS from [001]-oriented graphene, with the two components separated directly in the  $\ell = 0$  and  $\ell = \pm 1$  channels.

## References

- [1] V. Grillo, A. H. Tavabi et al. Nat. Comm. 8 (2017) 15536.
- [2] P. Schattschneider, S. Rubino et al. Nature. 441 (2006) 486–488.
- [3] E. Rotunno, M. Zanfrognini et al. Phys. Rev. B 100 (2019) 224409.
- [4] M. Zanfrognini, E. Rotunno et al. ACS Photonics 6 (2019) 620-627.