

New insights into optical property characterization of 2D semiconductor materials through time-correlated photon/electron spectroscopies

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Background:

The optical properties of luminescent materials are intricately tied to their structural and chemical characteristics at the nanometric, or even atomic scale. Understanding this relationship has long relied on the combination of scanning transmission electron microscopy (STEM) with external optical techniques such as absorption or photoluminescence spectroscopies, probing the excitation and emission mechanisms at the micrometric scale. However, the significant gap in spatial resolution between these methods and phenomena of interest often leads to ambiguous or even impossible interpretations of the measurements. Recent advancements in nano-optics seek to address this resolution disparity by enabling observation of luminescent material properties at nanometric scales. In that sense, STEM has undergone substantial instrumental and theoretical progress, providing valuable insights into optical properties through conventional inelastic electron scattering (electron energy loss spectroscopy, EELS) and photon emission (cathodoluminescence, CL) spectroscopies [1]. However, both techniques are hindered by spectral resolution limitations and the broadband excitation process of the electron beam. This contribution investigates the possibilities presented by the latest advancements in STEM techniques, specifically focusing on electron-photon time-correlations.

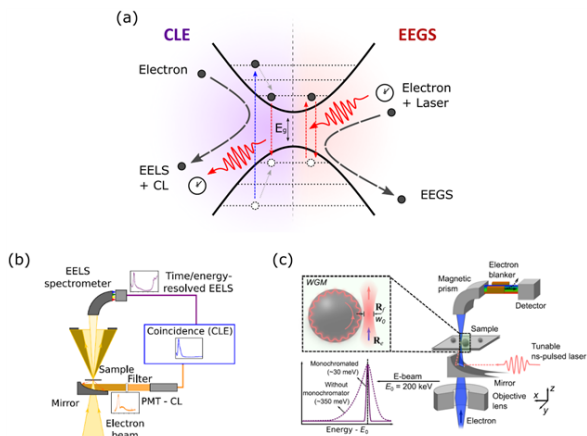
Methods:

To overcome these challenges, two techniques currently in development will be highlighted in this contribution:

- Cathodoluminescence Excitation Spectroscopy (CLE, Figure 1b), based on coincidence measurements between EELS and CL events. CLE offers insights into energy transfer pathways and excitation lifetimes at the nanometer scale, as previously reported [2,3].
- Electron Energy Gain Spectroscopy (EEGS, Figure 1c), an extension of the photon-induced near-field electron microscopy (PINEM) technique, investigates primary electron acceleration when the beam interacts with a strong optical field generated by a laser. By changing the energy of the photons, this technique can resolve ultra-fine optical resonances in the sub-meV range, revealing novel physical properties below the energy resolution achievable with state-of-the-art monochromated electron beams. So far, it has only been applied to photonic excitations, such as plasmons or surface polaritons [4].

Results and Conclusion: These techniques, with their distinct excitation and deexcitation processes (Figure 1a), are expected to be highly complementary, offering deeper insights into optical mechanisms at the nanometer scale. The present contribution focuses on using transition metal dichalcogenides (TMDs) as a model system of functional luminescent material to explore the insights provided by these correlated techniques.

Graphic:



Keywords:

Nano-optic, electron/photon spectroscopy, semiconductor TMD.

Reference:

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