From Quantum Confinement to Quantum Electrodynamics using nanoCathodoluminescence in a STEM

Mathieu Kociak¹

Cathodoluminescence (CL) has been used for decades to study luminescence of semi-conductors and presents obvious advantages with respect to Photoluminescence. This includes an easy access to high energies (UV regions and above), which is competitive for e.g high band gap materials study, or a strong absorption cross-section, interesting e. g. for the detection of diluted defects. Of course, an obvious advantage seems to be the spatial resolution that should be given by the sub-nanometre electron beams. However, this advantage used to fade away against physical realities that either degraded the spatial resolution (presence of an interaction pear, long charge carrier diffusion length...), the signal to noise ratio (low current in small electron probes, small collection angles, low figure of merit detectors...) or the relevance of the obtained data (saturation and non-linear effects at the electron currents necessary to get a decent signal to noise ratio). Thus, CL in high spatial resolution Scanning Electron Microscope (SEM) or Scanning Transmission Electron Microscope (STEM) has remained confidential in the past 20 years, despite promising pioneering works [1-3]. Now, the blossom of new luminescent objects (Quantum Dots, Quantum Wells, individual defects), with potential applications ranging from photovoltaics to quantum information, clearly urges the development of luminescence techniques with ultra-high spatial resolution. Indeed, all these objects for which the optical properties rely somehow on quantum confinement, are necessarily small (from a single atom to a few atomic planes).

I will try to show why we believe that a mixture of technological advances in machining and detectors, a new high efficiency STEM-CL design (nanoCL [4]), some unexpected reduction of the charge carrier diffusion length in heterostructured materials [4] and the somehow counterintuitive use of high voltages to *minimize* inelastic interaction is a game changer. I will review our recent use of nanoCL to study the quantum confinement at its relevant scale – a few nanometers. Beyond quantum confinement, even more fundamental but rather elusive quantum *electrodynamics* effects arise when studying quantum emitters. I will show, as exemplified on figure 1, that we have recently generated and characterized single photon states emission in a STEM [5] - pure quantum optical states with applications in quantum information.

This work, partially funded through ESTEEM2 (EU 7th Programme, GA 312483), has been mainly performed by the following persons that I want to warmly acknowledge: L. F. Zagonel, L. Tizei, Z. Mahfoud, S. Meuret, R. Bourrelier, A. Zobelli, M. Tencé and O. Stéphan with the support of the ORSAY STEM group.

References:

- [1] M. Grundmann, et al., Phys. Rev. Lett. 74, (1995) 4043.
- [2] U. Jahn, et al. Appl. Phys. Lett. 90, (2007) 161117.
- [3] H. P. Strunk, M. Albrecht, and H. Scheel, J Microsc 224, (2006) 79.
- [4] L. F. Zagonel, et al., Nano Lett. 11, (2011) 568.
- [5] L. H. G. Tizei and M. Kociak, Phys. Rev. Lett., 110 (2013) 153604.

^{1.} Laboratoire de Physique des Solides, CNRS/Univ. Paris XIII, Orsay, France

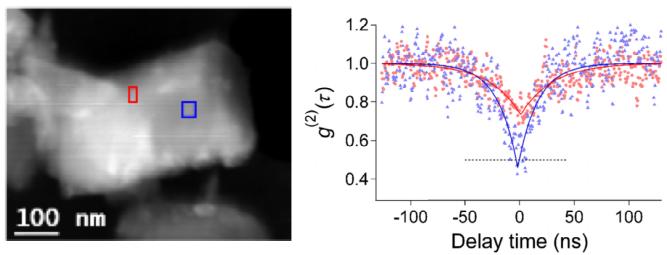


Figure 1. Measurement in a STEM of the quantum nature of light emission from individual Nitrogen-Vacancy centers in nanodiamonds nanoparticles. Left: high angle annular dark field image of a diamond nanoparticle. Right: time autocorrelation functions $(g^{(2)}(\tau))$ of the CL signal emitted when the electron beam is placed on two different area. The dip at zero delay is a signature of the quantum nature of the CL-emitted light. After [5].