High-spatial-resolution detection of new single-photon emission sources

Luiz H. G. Tizei, Sophie Meuret, Romain Bourrellier, Anna Tararan, Odile Stéphan, Mathieu Kociak, and Alberto Zobelli

A scanning transmission electron microscope coupled to a Hanbury Brown and Twiss interferometer is used to achieve cathodoluminescence in a novel experimental technique.

There is currently a large scientific effort being made to identify, characterize, and control new single-photon emission sources (SPEs), with the aim of extending their spectral range. SPEs—light sources that are emitted as single particles or photons—are intrinsically small and, to a first approximation, are necessarily a two-level system. Furthermore, the typical size of active structures in modern light emitters is in the nanometer range (e.g., for quantum wells) or smaller (for point defects). The identification of new SPEs therefore requires a technique that permits high spatial resolution and optical characterization. Although pure optical or scanning probe techniques can be used to achieve high spatial resolution and optical characterization of SPEs, it is not possible to meet both these requirements with a single technique.

It is thought that hexagonal boron nitride (h-BN) is a promising candidate for extending the spectral region of available SPEs into the far-UV (presenting an excitonic emission at 5.8eV). Moreover, common h-BN samples produce more complex emissions than have generally been attributed to the presence of structural defects. Despite a large number of experimental studies, it has not yet been possible to attribute specific emission features for the proper identification of defective structures. Light emission from these structural defects occurs at extremely localized regions, with lateral sizes of about 80nm and the localized regions indicate the possible existence of single defects. The large spatial distribution is attributed to charge diffusion caused by the lack of potential barriers. The occurrence of single-photon emission at these sites has been confirmed by light intensity interferometry in an electron microscope.

In our work, we have taken a different (i.e., not purely optical) route to obtain high spatial resolution and optical characterization of SPEs simultaneously. In our approach we use a focused 1nm-wide fast electron beam in a scanning transmission electron microscope (STEM) that is coupled to a Hanbury Brown and Twiss (HBT) interferometer. The electron beam excites the sample in very small regions, which leads to light emission—cathodoluminescence (CL)—and allows optical characterization at the nanometer scale. By analyzing the light output with...
an HBT interferometer, it should be possible to demonstrate the existence of an SPE.

Our experimental setup is illustrated in Figure 1. We use the focused electron probe of our STEM (VG HB501), operated at 60kV, as the excitation source. The light emitted from a thin sample is captured by a collection system that we have built in-house. In this system, we use a parabolic mirror to transport the light out of the microscope’s vacuum. The resulting output can then be analyzed with either an optical spectrometer (to gather spectral information about the excited structure) or the HBT interferometer (for SPE detection).

Using this same experimental design, it has previously been demonstrated\(^7\) that the emission spectra of gallium nitride quantum disks separated by 5nm of aluminum nitride in nanowires can be distinguished. In fact, the spectral resolution that can be achieved is not limited by the size of the electron probe, but rather by the diffusion of charge carriers in the material. We show an example of typical data from such a system in Figure 2(a). We obtained this data by scanning the electron beam over the sample in a 2D array of points and by acquiring one spectrum at each point to form a spectrum image (SPIM). The typical integration time for an individual spectrum is 10ms. A SPIM acquisition (tens of thousands of spectra) therefore generally lasts a few minutes.

With the high spatial resolution of our CL spectrum imaging, and the capability of STEMs for imaging individual atoms and defects, it should be possible to detect SPEs at high spatial resolution. Indeed, we have recently shown that with our experimental setup (i.e., STEM-CL and HBT interferometer) we can detect a well-known SPE (the nitrogen vacancy center in diamond nanoparticles).\(^8\) We measured the second-order correlation function, \(g^{(2)}(t)\), with anti-bunching, at a spatial resolution of about 150nm, as shown in Figure 2(b). We have also recently used the same experimental setup to demonstrate the possibility of measuring the lifetime of an ensemble of identical SPEs. We achieve this by measuring the width of the bunching peak (a high correlation of photons at time zero). This effect, which only occurs for samples excited by electrons,\(^9\) thus opens up the possibility of nanometer-resolution lifetime mapping.\(^10\)

In summary, we have demonstrated the possibility of detecting single-photon emission sources from different materials with a novel experimental setup. In our experimental design, we use a scanning transmission electron microscope (to produce cathodoluminescence) coupled with an HBT interferometer. The high spatial resolution of our technique also allows the identification of regions with specific defects of structures in heterogeneous materials, even if they are confined to sub-micrometer areas. In our future work, we plan to continue searching for new SPEs in different materials, including 2D monolayers. Moreover, we hope to link different spectro-microscopy techniques with atomically resolved imaging to identify the atomic structure of such SPEs.

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Author Information

Luiz H. G. Tizei, Sophie Meuret, Romain Bourrellier, Anna Tararan, Odile Stéphan, Mathieu Kociak, and Alberto Zobelli
Laboratory of Solid State Physics
University of Paris-Sud, CNRS
Orsay, France

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