Resolving semiconductor carrier dynamics at the nanoscale through photon-pump electron-probe microscopy

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The performance and efficiency of photodetectors, photovoltaics, and other solid state devices are critically influenced by the dynamics of free carriers [1]. For nanoscale devices, techniques are required to visualise these dynamics in space and time with picoseconds or lower temporal resolution and spatial resolution far below the optical diffraction limit. Pump-probe techniques, where the sample is pumped with a laser pulse and probed with a nanometre sized electron pulse, have been shown to visualise carrier dynamics on bulk surfaces [2]. The optical excitation of carriers gives rise to a local, transient contrast in the secondary electron (SE) signal induced by the probing electron pulse in a scanning electron microscope (SEM). However, in these past experiments the high spatial resolution of the electron beam could not be fully taken advantage of due to the massively larger laser focus, measuring tens of microns and thereby limiting the spatial resolution [2].

Our setup, consisting of a FEI Quanta 200 SEM equipped with an inverted light microscope below the sample, enables the use of high-NA optical objectives for optical imaging and excitation [3]. This allows us to tightly focus a Coherent Vitara-T Ti:Sapph femtosecond laser beam into a 700 nm spot and scan the electron beam in and around the laser focus area. Compared to previous work, this is over an order of magnitude improvement in spatial resolution [2]. By means of dispersion compensating optics, the optical pulse duration can be limited to 15 fs, meaning that the laser excitation is well defined in space as well as time. Moreover, the probing electron beam can be pulsed by means of beam blanking technology yielding an electron pulse duration of 90 ps with a spatial resolution well below the optical diffraction limit [4].

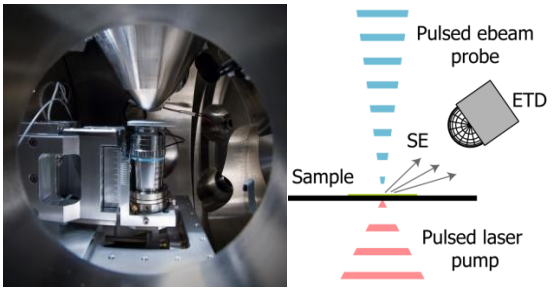

Figure 2 The FEI Quanta 200 SEM based setup accommodates an inverted optical microscope underneath the sample, meaning that the specimen can be the subject of simultaneous electron and high NA optical imaging.

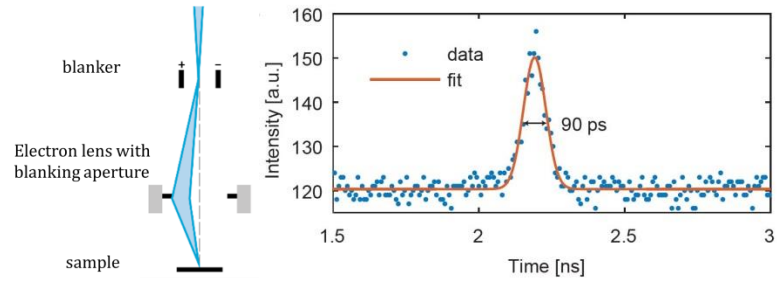

Figure 3 The electron beam is pulsed using beam blanking technology, enabling a 90 ps pulse duration.

We will show proof of principle pump-probe measurements on semiconductor nanomaterials with the goal of mapping carrier diffusion with sub-micron spatial resolution and sub-ns temporal resolution. A time dependent change in SE yield is observed when the delay between laser pump and electron

probe pulses is varied, and the decay time of this contrast is in line with literature values of the carrier lifetime.

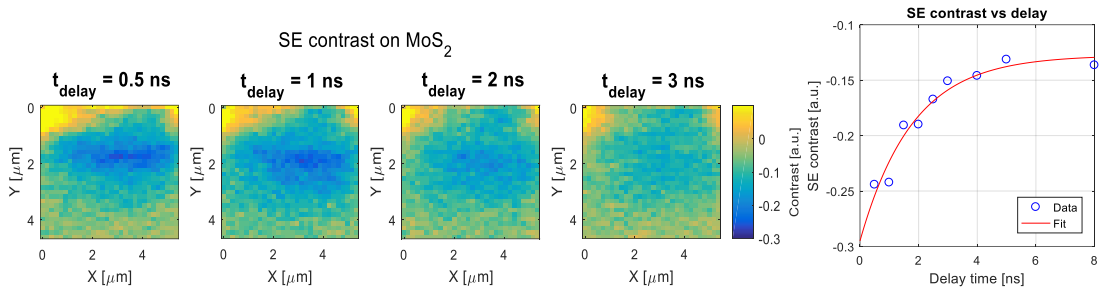

Figure 4 Proof of principle pump probe measurements on MoS2. The laser induced SE yield images on the left indicate that laser excitation results in a reduction of the SE yield. An exponential fit shows a restoration time of 1.8±1.1 ns.

[1] J. Sun et al., **J. Phys. Chem. Lett. 7(6), 985-994 (2016)**.

[2] O.F. Mohammed et al., **ACS Appl. Mater. Interfaces 9(1), 3-16 (2017).**

[3] A.C. Zonnevylle et al., **J. of Microscopy 252, 58 - 70 (2013)**.

[4] R.J. Moerland et al., **Opt. Express 24(21), 24760-24772 (2016)**.