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Mapping Giant Oscillator Excitons in Semiconducting Nano Wires

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Controlling exciton emission is an important step towards new optical devices. Monochromatic electron energy-loss spectroscopy (EELS) can probe these quasi-particle excitations from the near infra-red to the ultraviolet spectral regions with sub-nanometer spatial resolution. The much-improved energy resolution reduced tails of the zero-loss peak make this energy region accessible.

To compare EEL spectra from different instruments and materials, the features are quantified to an absolute scale. Least square fitting is used to model the spectrum including the zero-loss peak. This results in the exact position in energy and scattering intensity of each feature. The instruments used in this study are the Zeiss Libra MC 200 with a spatial resolution of 240 pm and an energy resolution better than 150 meV. A Gatan cooling holder can be utilized for cryo-EELS. The MAC-STEM at ORNL provides superior spatial (78 pm at 100 kV) and energy resolution (11 meV).

The small dimensions of semiconductor nanowires allow them to be investigated with optical spectroscopy and EELS without requiring additional specimen preparation steps. Figure 1 shows a GaAs-AlGaAs-GaAs core shell nanowire overlaid with a map of the exciton signal. This signal is about 10000 times larger than the volume plasmon and its intensity (area under the peak is about 100 times stronger than a Wannier-Mott exciton in GaAs. This exciton signal comes from the outer 20 nm of the nano wire shell, which consists of heavily defective GaAs. An atomic resolution high angle annular dark field image from a Nion UltraSTEM200, shows that the surface consists of less than a nanometer of amorphous film but appears otherwise completely crystalline.

The exciton signal appears only in the first 20 nm of the GaAs layer. The signal is extremely strong and sharp and is identified as a Frankel exciton originating from point defects. The exciton is visible at room-temperature which is also consistent with the high binding energy of a Frankel exciton. The EL2 defect in GaAs has the same energy as the observed excitation.[1] This EL2 defect is an arsenic anti-site defect combined with a vacancy.[2] The map in Figure 1 is therefore a direct map of this exciton.

Similar excitons have been found in a variety of relevant semiconductor materials (Figure 2).[3] These unusually strong excitations can therefore be used to study point defects in these materials. These excitations are associated with a mid-level gap state and therefore are important for the efficiency of electronic and opto-electronic devices.

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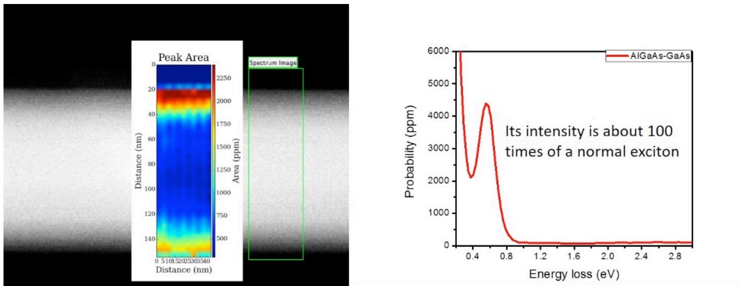
[4] The authors acknowledge funding from ARO grant W911NF-13-1-0428. Work done at ORNL was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division.



	Position(eV)	Width(eV)	Area (ppm)
AlGaAs-GaAs	0.617	0.158	1059

Figure 1. HAADF image and exciton map of a GaAs-AlGaAs-GaAs core shell nanowire with the spectrum from the exciton on the right.

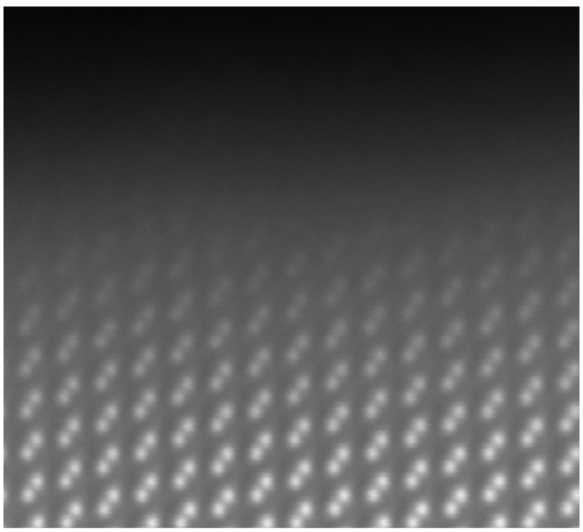
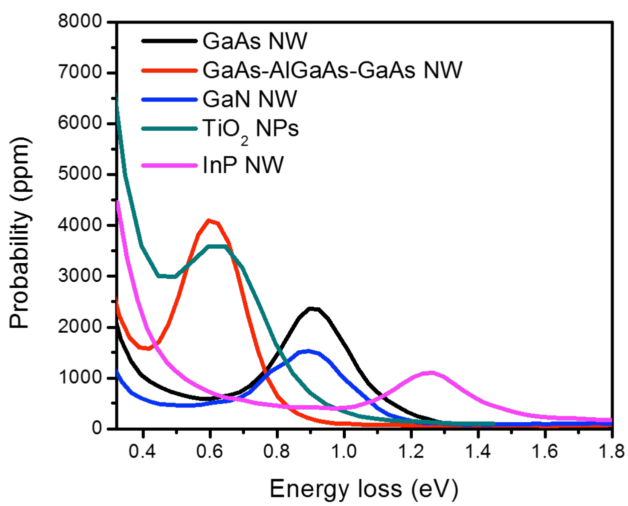


Figure 2. HAADF image of the GaAs surface of the GaAs-AlGaAs-GaAs core shell nanowire showing perfect crystallinity and thin amorphous surface. Dumbbell features are separated by 0.14 nm.



Material	Position(eV)	Width(eV)	Area(ppm)
GaAs	0.90	0.23	885
GaAs-AlGaAs-GaAs	0.62	0.16	1059
GaN	0.90	0.25	631
TiO_2	0.64	0.23	1254
InP	1.26	0.37	569

Figure 3. Excitons from various semiconductor nano wires with table of excitation energy, lifetime broadening, and scattering intensity.

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