Controlled integration of nanocrystals on the surface of group III-nitride light-emitting epitaxial heterostructures

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Currently an extensive range of colloidal nanocrystals (NCs) can be readily produced by diverse chemical methods. These nanomaterials present novel physical properties and can be regarded as building blocks to the nanofabrication of smaller, energy efficient and faster devices. However to form a functional device with reasonable complexity, these nano-building blocks have to be conveniently arranged and connected. This requires incorporation into an interconnectable, electrically and/or optically active framework. Such endeavour is a key challenge in nanoscience and nanotechnology. Here we address this via a novel and inexpensive approach that exploits the spontaneous formation of nanoscale inverted hexagonal pits (IHPs) at the surface of In-containing III-nitride epitaxial heterostructrures.

We demonstrate that these morphological features, which can be designed with a suitable size, depth and density, can effectively organize and control the incorporation of NCs at the surface of efficient light emitting devices. Nanoengineering of the relative sizes of NCs and IHPs allows the number of NCs incorporated into each hexagonal pit to be controlled precisely.

Although the selective incorporation of virtually any kind of NCs into the IHPs is possible due to interfacial capillarity forces acting during the deposition method, as a proof of concept, this work will focus on the nanomanipulation of Au nanocrystals with sizes ranging from 30 to ~5 nm. It will be shown that each nano-pit can accommodate either individual NCs, or well-defined "clusters" of NCs. Controlled self-assembly, with areal densities up to $10^9/cm^2$, was achieved with precisions below the limits of conventional lithographic techniques, over macroscopic length scales.

The successful integration with nanometer-scale control of colloidal NCs onto the surface of III-nitride semiconductor heterostructures will bring together the benefits of *bottom-up* and *top-down* approaches towards the creation and control of nanostructures. As complementary realms of activity, the combination of both methodologies will certainly contribute to the achievement of novel functional nanodevices, and also give new scientific insights of a more

fundamental kind. In this context, new properties may arise through the exploitation of cross-coupling effects, namely by the unrivalled versatility of wet chemistry to produce a wide range of nanomaterials, to be combined with the ease of charge injection and the high efficiency of light emission of group III-N bipolar devices. Such synergetic incorporation also opens the way to investigate and tailor the physical properties of elementary NC clusters, down to isolated individual NCs, as well as the coupling of elementary excitations (e.g. electronic, photonic and plasmonic) between the nitride framework and the nanoscale building blocks.

References:

S. Pereira, Tito Trindade, Manuel A. Martins, Ian M. Watson, Diane Zhu and Colin J. Humphreys "Controlled integration of nanocrystals in inverted hexagonal nano-pits at the surface of light-emitting heterostructures" *Adv. Mater.*, **2008**, *20*,**-cover story-**

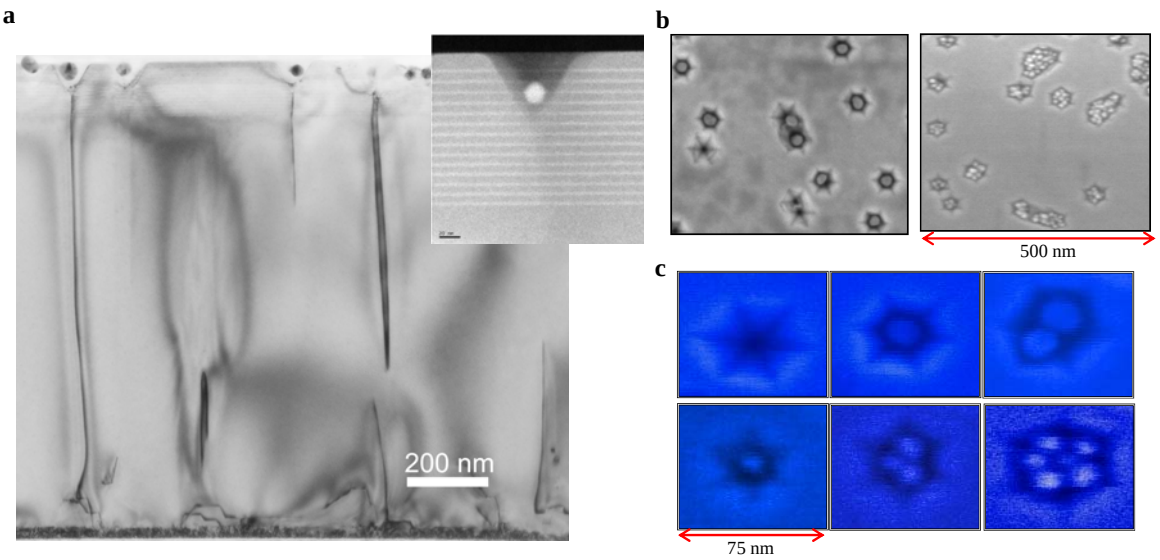

Figure 1: **Cross section TEM and plane view SEM images showing effective selective incorporation of Au NCs into the IHPs.**

a) Bright field TEM micrograph showing the nitride heterostructure from the sapphire interface up to the surface, where the Au NCs are incorporated inside the IHPs. The higher magnification inset shows a STEM-HAADF micrograph of the InGaN/GaN MQW region near a pit. An isolated Au NC can be observed in detail. The 14-wells forming the MQW can also be observed. In this TEM imaging mode the contrast is strongly dependent on the atomic number Z of the atoms encountered by the incident probe. The Au NC is the brightest feature (Z_{Au}=79) while the InGaN well regions also appear brighter than the GaN barriers and cap layer (Z_{In}=49, Z_{Ga}=31).

b) Plane view SEM micrographs demonstrating selective incorporation of NC inside the Pits for Au nanoparticles with sizes of ~30nm (left) and ~5 nm (right).

c) The upper panel shows an empty nano-pit in detail, a ~30 nm Au NC inside a IHP and a NC dimmer. The lower panel shows NC assemblies with increasing complexity using ~15 nm NCs; namely 1 NC per pit, 4 NCs (only 3 visible) forming a tetrahedron and finally a 3-monolayer NC "lattice" with 5 NCs visible at the surface.