X-ray excited optical luminescence in InAs-based nanostructures: preliminary results

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Because of their versatile applications, the past decades have witnessed rapid growth in research and development of luminescent materials.[1] However, further progress towards advanced optical devices with higher efficiencies, lower costs, and longer lifetimes are based on nanotechnology (e. g., nanopowders, nanostructures, quantum wells/wires/dots, nanoparticles, etc.).[2] All together requires a better understanding of the role played by confining effects, interfaces and surfaces, defect formation on the optical properties. As a result, there have been strong motivations for the application of new optical characterization techniques using several probes like photons, electrons, ions, and also x-rays. Many studies have been carried out in almost all the fields: photoluminescence, cathodoluminescence, electroluminescence, thermoluminescence, bioluminescence, among others.[3] New optical phenomena in known materials have become possible to observe: induced dichroism, depolarization of luminescence with increasing excitation power, two-photon absorption, luminescence at forbidden transitions, hot luminescence, biexcitonic emissions, electron-hole drops, ultrafast processes, self-focusing of light, non-linear phenomena, etc.[4] The range of luminescent compounds has been also broadened owing to the production of complex systems, and often, materials that do not exist in nature at all, i.e., artificial atoms, biomolecular electronic compounds, quantum structures, stressed layers and superlattices.[1]

However, there are still new light detection strategies to overtake the next generation of advanced optical devices. In the present work, we propose a new experimental approach based on the simultaneous application of different non-destructive techniques at the hard X-ray microprobe of the European Synchrotron Radiation Facility (ESRF).[5] This alternative system is designed for parallel collection of multiple signals: scanning μ-X-ray fluorescence, X-ray absorption spectroscopy (XANES/EXAFS) and scanning μ-X ray-dispersive photoluminescence. The synchronous detection of different emission processes on the sub-micron scale provides valuable insights into the elemental composition of the crystal lattices and defects contained within the nanostructures.[6] Accordingly, a mini-cryostat has been specially designed with optimal optical access, broad temperature range (11-300 K), excellent thermal stability ($\Delta T/T \leq 2.3 \times 10^{-2}$ at 12 K) and extremely short working distances (4.5 mm).[7] Given the ID22 beamline capabilities with achromatic micro-focusing optics (up to 18 keV), multi-element Si(Li) detector and high photon flux (10^9 - 10^{12} ph/s), a wide range of excitation/relaxation pathways could be explored and a few elemental monolayers probed. We have applied this new technique to the optical study of molecular beam epitaxy (MBE) grown heterostructures: InAs quantum wires (1.6 ML thick) on (001) InP substrates and InGaAsSb quantum dots (2.5 ML thick) on (001) GaAs substrates. Their luminescence, basically in the near IR optical range, presents astonishing properties due to quantum size effects, which potential applications in optical fiber telecommunications or as single photon emitters or resonant tunneling structures for high-frequency. By exciting with the X-ray microbeam we are able to collect not only quantum luminescence but also optical signal from the barrier material and substrate. Through the latter, no heating induced by higher X-ray photon flux is

observed on the different samples. In fluorescence detection mode, EXAFS spectra from the InAs quantum wires at the As K-edge are detected with orientation-selective excitation (the electric field vector of the synchrotron radiation parallel and perpendicular to the quantum wires).

In summary, the development of the X-ray excited optical luminescence technique within the framework of the hard X-ray microprobe offers unique and complementary information on the short/long-range order and electronic structure of crystalline nanostructures directly involved in the light emission processes. In contrast to conventional XANES and EXAFS analyses, we will be not only element selective but also site selective since the wavelength of the luminescence may depend on the elemental local environment. By measuring in both fluorescence and luminescence yield modes at the same time, different sample depths can be probed due to the shorter attenuation length of the visible photons.

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