PROPOSAL CODE HC-3808

THREE-DIMENSIONAL BRAGG COHERENT X-RAY DIFFRACTIVE IMAGING OF A SINGLE GE/SIGE NANOPARTICLES PRODUCED BY FEMTOSECOND LASER PRINTING

Semiconductor and dielectric nanostructures attract increasing attention of researchers due to their high potential as a substitution for plasmonic metal-based metamaterials. The latter suffer from intrinsic Ohmic energy losses at optical frequencies, which considerably limit the device efficiency. In contrast, so-called all-dielectric metamaterials have much lower absorption losses, and, moreover, can exhibit strong Mie-type resonances in the optical response, which is favourable for the realization of the fundamental task of nanophotonics – light manipulation at the nanoscale [1]. Recently introduced femtosecond laser printing technique was further developed for the fabrication of crystalline single Ge and SiGe nanoparticles [2]. Individual Ge nanoparticles (NPs) act as nanoresonators whose response is determined by their size and shape. One of the recently introduced techniques for the fabrication of nearly ideally spherical NPs is known as femtosecond (fs) laser printing [3]. Ge and SiGe NPs also exhibit Mie-type resonances in the visible light scattering spectra which make them promising candidates for needs of nanophotonics [4]. Thus, the demonstrated fs laser printing of size-controlled Ge NPs provides new possibilities for the fabrication of novel metamaterials.

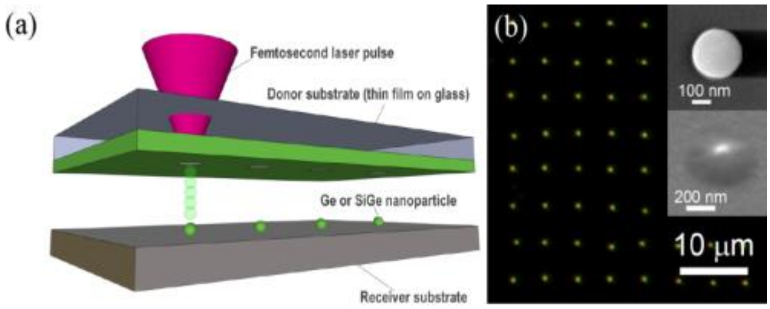

Figure 1. (a) Scheme of the fs laser printing method of NPs; (b) Dark-field microscopic and SEM images of a Ge NP array printed by 400 nm, 7 nJ single fs laser pulses.

The crystallinity and strain information was probed by coherent X-ray diffraction with the aim to apply Bragg coherent x-ray diffractive imaging technique. The detailed studies of these structures will provide an insight into the structure-functionality relation in nano-photonics. Additional, we develop approaches for a full characterization of nano-crystalline structures with a limited a priory information.

Objectives for the studies with respect to the deposition was: determination of crystallinity by X-ray diffraction; determination of a preferred orientation with respect to the receiver substrate; strain distribution within single particles, leading to altered SHG efficiency; mapping of defects such as stacking faults, dislocations, inclusions due to the deposition process.

We have employed in-situ BCDI technique to study a single Ge NP. The measurements were performed at ID01 beamline of ESRF with height resolution sample scanner and 2-circle detector arm. We have prepared well-ordered arrays of Ge NPs (see Fig.1 (b)). The regions of interest were marked with FIB which simplified sample alignment and data acquisition. We used a coherent x-ray beam with a size of about 0.4x0.4μm^2 to fully illuminate a single NP. Experimental scheme with two detector position was employed, one in the near-field for Bragg peaks alignment and the second for far-field measurements at a distance of a two meters downstream the sample at double Bragg angle. With this setup the interference fringes related to a finite size of the particle were well resolved. Single 111 reflections were found from NPs produced by different laser energy. We recorded rocking curves in the vicinity of Bragg angle in the range of ±1 degrees to obtain 3D reciprocal maps of selected NPs.

Bragg reflections were measured from the NPs produced by fs laser with energies in the range of 72 to 75 mW. BCDI data obtained from different energy groups revealing single and poly-crystalline structures

(Fig.2). Therefore, we divide the cases of single-crystal or polycrystalline NPs depending on the formation conditions. This information will contribute to the understanding of the influence of the crystal structure of the NP on its optical properties, in particular, on the second harmonic generation. Additionally, we address an issue of NPs rotation under high-flux x-ray beams.

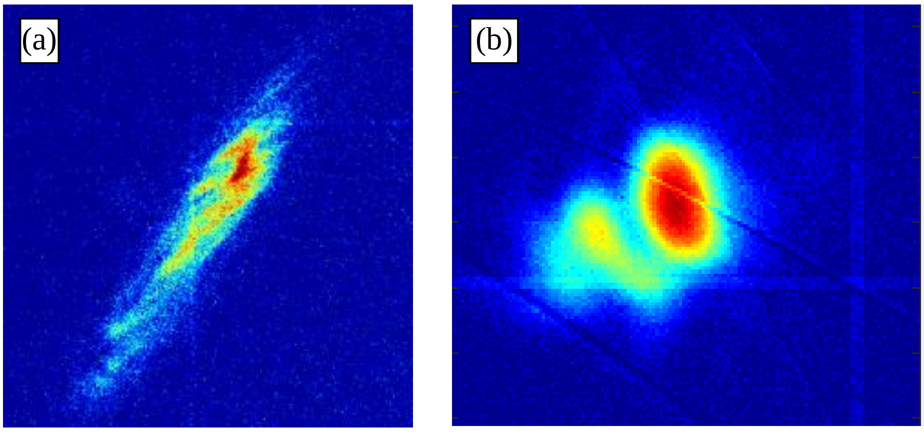

Figure 2. Experimental far-field coherent diffraction patterns from polycrystalline (a) and single-crystalline NPs (b) around (111) reflection

We found that free-standing nanoparticles can change their orientation under the synchrotron beam irradiation. This effect led to go out the nanoparticle from the Bragg diffraction conditions during scanning around the reciprocal lattice point. Ultimately, this effect prevented the receipt of complete information for reconstructing the structure of a single nanoparticle. It should be noted here that this problem will be key point in high-bright sources: upgrade to EBS and X-ray free-electron lasers. Particular attention should be paid to the method of immobilization of free-standing nanoparticles (not only semiconductor, but also organic, including single biomolecules) for conducting CDI experiments. We propose the use of a single-walled carbon nanotube (SWCNT) free-standing membranes to immobilize semiconductor nanoparticles in future experiments.

The ability of BCDI to reveal a fine information on the structure and strain within single NPs would pave the way for deeper understanding of their crystallization kinetics and physics of formation by fs laser printing method. The obtained knowledge will be taken into account for the upcoming fs laser printing experiments with new materials. As an intermediate result an issue of the nano-samples fixation is a crucial problem to be solved. Especially at the scales of 100 nm and when the particles are deposited on a foreign substrate. Further tests will lead to more outcome from Bragg coherent x-ray diffractive imaging experiments.

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