

IN-SITU MONITORING OF LASER ANNEALING BY MICRO-RAMAN SPECTROSCOPY FOR HYDROGENATED SILICON NANOPARTICLES PRODUCED IN RADIO FREQUENCY GLOW DISCHARGE

D Hill, T Jawhari¹, J Garcia Cespedes, J Alvarez², E Bertran

FEMAN group, Departament de Física Aplicada i Òptica, Universitat de Barcelona, Martí i Franqués, 1, 08028 Barcelona, Spain

¹Serveis Científico-tècnics, Universitat de Barcelona, C/ Lluís Solé i Sabarís, 1-3, 08028 Barcelona

*²Centre de Recerca i Investigació de Catalunya, S.A. C/ Margalló, 12 08017 Barcelona
daniel.hill@ub.edu*

Plasma enhanced Chemical Vapour Deposition (PECVD) has been used to produce amorphous hydrogenated Si nanoparticles (Fig. 1) of high purity from square wave modulated radio frequency (rf) glow discharges [1, 2] across the flow of argon diluted silane. The samples deposited on glass microscope slides and TEM grids were of various size distributions and structures, as determined by the process parameters. Chemical characteristics, structure and other information associated with the formation of particular peaks were then determined through the use of Micro-Raman Spectroscopy. Earlier Raman studies showed particular structural and compositional characteristics of silicon nanoparticles produced by diverse synthesization techniques based on rf glow discharge, electrochemistry, ball-milling, reactive magnetron sputtering and by pulsed laser ablation [3-7].

Micro-Raman back scattering measurements of different coverages of hydrogenated silicon nanoparticles were performed at various CW powers (Fig. 2). We present a study of the changing structure of these samples due to the induction of nanocrystallites by the Ar Ion laser when taking the Raman Spectra. As the power of the laser is increased the Si-Si bond vibrational intensity in the Raman spectra also increases with the peak narrowing and shifting to a higher wave numbers which we interpret as further crystallisation taking place as further annealing of the Si:H surfaces. As expected, the intensity is higher for areas with greater coverage of Si. High resolution transmission electron microscope (HRTEM), electron energy loss spectroscopy (EELS) and selected area electron diffraction (SAED) analysis confirmed the structural changes of particles, which were associated with the increase of temperature, surface oxidation and hydrogen diffusion induced by laser annealing at atmospheric conditions.

References:

- [1] J Costa, G Sardin, J Campmany and E Bertran, *Preparation of nanoscale amorphous silicon based powder in a square-wave-modulated rf plasma reactor*, Vacuum, **45** (1994)1115-1117.
- [2] Lloret, A.; Bertran, E.; Andújar, J.L.; Canillas, A.; Morenza, J.L., *Ellipsometric study of a-Si: H thin films deposited by square wave modulated rf glow discharge*, Journal of Applied Physics, **69** (1991)632-638.
- [3] S. Huet, G. Viera and L. Boufendi, *Effect of small crystal size and surface temperature on the Raman spectra of amorphous and nanostructured Si thin films deposited by radiofrequency plasmas*, Thin Solid Films, **403-404** (2002)193-196
- [4] F. M. Liu, B. Ren, J. H. Wu, J. W. Yan, X. F. Xue, B. W. Mao and Z. Q. Tian, *Enhanced-*

Raman scattering from silicon nanoparticle substrates, *Chemical Physics Letters*, **382**(2003)502-507

[5] C. Araujo-Andrade, F. J. Espinoza-Beltrán, S. Jiménez-Sandoval and J. González-Hernández, *Synthesis of nanocrystalline Si particles from a solid-state reaction during a ball-milling process*, *Scripta Materialia*, **49** (2003)773-778.

[6] Y. Leconte, P. Marie, X. Portier, M. Lejeune and R. Rizk, *Combined growth of Si nanoparticles and crystallized silicon layers at 200 °C by reactive magnetron sputtering*, *Materials Science and Engineering B*, **101**(2003)194-198.

[7] Takafumi Seto, Takaaki Orii, Makoto Hirasawa and Nobuhiro Aya, *Fabrication of silicon nanostructured films by deposition of size-selected nanoparticles generated by pulsed laser ablation*, *Thin Solid Films*, **437** (2003)230-234

Acknowledgements:

This study was partially supported by the *Generalitat de Catalunya* (2001SGR 00078), the *CICYT* of Spain, project MAT 2002-04263-C04 and COOP-CT-2004-513275 project of the European Community. The authors thank Serveis Científico-tècnics of the Universitat de Barcelona (SCT-UB) for measurement facilities.

Figures:

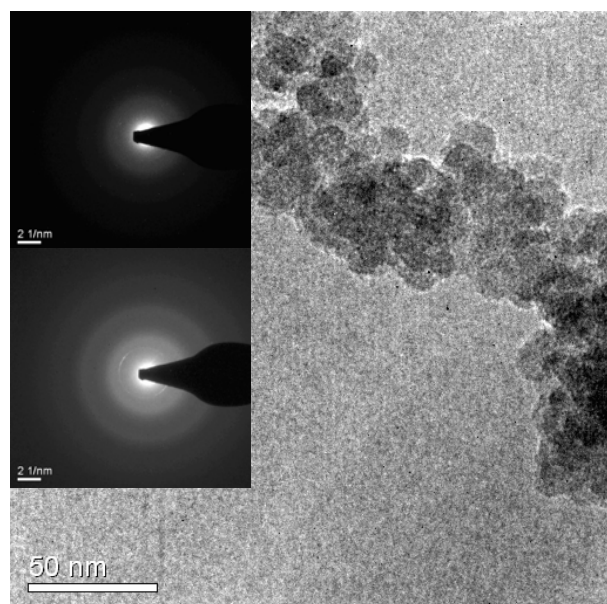


Figure 1: Main Image: TEM micrograph showing size, relative density and morphology of the Si:H nanoparticles. Upper Inset: SAED image taken far from laser incidence showing the amorphousness of the hydrogenated Si nanoparticles. Lower Inset: SAED image taken on edge of laser incidence, showing some structure of $\sim 3\text{\AA}$.

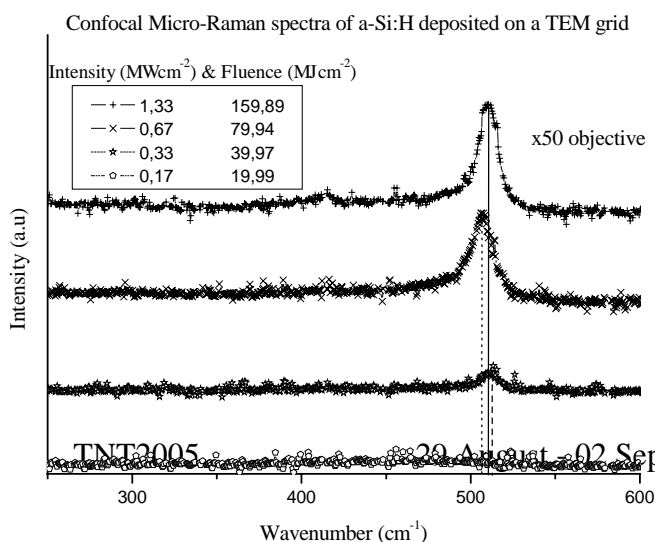


Figure 2: Raman spectra of the hydrogenated Si nanoparticles, with $>2\text{mm}$ separation between lased areas. The peak

at $\sim 500\text{cm}^{-1}$ corresponds to the vibration of the Si-Si bond in Si nanocrystallites.