

Manjunath Balagopalan¹

Gloria Conte¹, Omid. R. Ranjbar-Naeini¹, Yani Chen¹, Catalina C. Benejam², Miguel Pruneda², Oili Ylivaara³, Jouni Ahopelto³ and Clivia M. Sotomayor Torres¹

¹ INL-International Iberian Nanotechnology Laboratory, Avenida Mestre José Veiga s/n, 4715-330 Braga, Portugal

² Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and BIST, Campus UAB, 08193 Bellaterra, Spain

³ VTT – Technical Research Centre of Finland Ltd, Finland

manjunath.balagopalan@inl.int, clivia.sotomayor@inl.int

Grain-boundaries in nanocrystalline silicon: Insights from Raman Spectroscopy

Nanocrystalline silicon (nc-Si), composed of nanoscale crystallite grains, has garnered significant attention for decades due to its distinct properties compared to single-crystalline silicon. nc-Si offers a versatile affordable alternative to, e.g., silicon-on-insulator, enabling applications in MEMS, photonics and owing to its uniqueness concerning tunable mechanical optical and thermal properties associated with the presence of nano crystallites and its compatibility with established silicon processing methods. Recent studies demonstrated that optomechanical devices based on nc-Si can achieve higher quality factors than their single-crystal counterparts, which could be attributed to its features such as midgap states possibly arising from the grain boundaries (GB) [1,2]. Therefore, a comprehensive understanding of the structural and physical properties of nanocrystalline silicon, particularly the role of grain boundaries, is critical to establish structure-property relationships and optimise the material for a wide range of technological applications. In this work, Raman spectroscopy was used as a powerful characterisation tool to investigate the structural and physical properties of nanocrystalline silicon. The nc-Si samples, typically prepared by annealing a layer of amorphous silicon, were investigated using polarized Raman scattering and were found to be highly isotropic (~95%). This pronounced isotropy is attributed to the random orientations of nanocrystalline grains. High-resolution Raman scattering investigations revealed additional vibrational bands, which originate from the grain-boundary regions. Their assignment was further corroborated by density functional theory (DFT) calculations of the local vibrational modes associated with the GB structure [3]. Furthermore, to elucidate the role of GB in thermal transport, the thermal conductivity (κ) of nanocrystalline silicon (nc-Si) was investigated using the contactless two-laser Raman thermometry [4], which offers high spatial resolution and low measurement uncertainty compared to conventional contact methods. The thermal conductivity was extracted by analysing the spatial temperature decay in steady-state conditions using Fourier's law. For 220 nm thick membranes, nc-Si exhibits $\kappa \sim 70 \pm 2 \text{ Wm}^{-1}\text{K}^{-1}$, lower than that reported for crystalline silicon ($80 \pm 3 \text{ Wm}^{-1}\text{K}^{-1}$) films of comparable thickness [4]. The observed reduction in the thermal conductivity of nc-Si is due to the strong phonon scattering at GB inherent to the nanocrystalline microstructure, with further contribution from the phonon confinement effects within the nano-grains.

References

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