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The Effects of Deposition and Processing Parameters on the Electronic Structure and Photoluminescence from Nitride-Passivated Silicon Nanoclusters

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Silicon nanoclusters (Si-ncs) exhibit unique optical properties as a result of quantum confinement effects. In particular, efficient luminescence from Si-ncs has driven research aimed at understanding the behaviour of these materials due to their potential for the development of Si-based light sources. Commonly, Si-ncs are formed within a Si-based dielectric. Difficulties arise in the understanding of these materials due to effects related to the Si-nc/dielectric interface, as well as due to the physical properties of the dielectric matrix. This situation is further compounded by fabrication specific issues, where the use of different deposition systems or source gases for the fabrication of Si-nc containing thin films can alter the observed optical behaviour of the materials.

Here, we compare and discuss Si-nc embedded silicon nitride samples fabricated using three different chemical vapour deposition (CVD) systems: plasma-enhanced CVD (PECVD), inductively coupled plasma CVD (ICP CVD), and electron cyclotron resonance PECVD (ECR PECVD). For each system, the sample compositions were controlled through the variation of the deposition gas flow rates. Post-deposition the samples have been subjected to thermal annealing in a quartz tube furnace under flowing Ar, Ar + 5%H, N, or N +5%H. The characteristics of the Si-ncs are strongly dependent on both deposition and processing parameters, as evidenced by variations in their measured luminescent properties and electronic structure. Photoluminescence (PL) experiments were conducted using a HeCd laser emitting at 325 nm as the excitation source. X-ray absorption spectroscopy (XAS) and x-ray excited optical luminescence (XEOL) experiments were conducted at the spherical grating monochromator (SGM) and variable line spacing parallel grating monochromator (VLS PGM) beam lines of the Canadian Light Source synchrotron facility.

PL measurements demonstrate that for each deposition system the peak emission wavelength for a sample can be controlled over the entire visible region of the electromagnetic spectrum through the variation of the gas flow rates used during the deposition, with the peak emission wavelength increasing with the index of refraction (which scales with an increase in silicon content). The PL intensity was found to depend on the temperature and ambient gas used in the annealing process. While the trends in the peak emission wavelength were similar between deposition systems, the ICP CVD deposited samples exhibit a shoulder on the shorter wavelength side of the main peak. Distinct emission behaviour has also been observed for nitrogen-rich and silicon-rich samples. While the origin of the luminescence from nitrogen-rich samples is expected to be defect-related rather than from Si-ncs, the emission intensity is on the same scale as that observed for silicon-

rich samples. Furthermore, the peak emission wavelength in the nitrogen-rich samples has also been found to be tunable through the deposition gas flow rates.

XAS performed at the Si *K*-edge provides evidence of increased Si-Si bonding in as-deposited samples of higher excess silicon concentrations from each CVD system, as seen in Figure 1. The results can be correlated with increased Si-nc size and the shift in peak PL emission wavelength observed. Similarly, an increase in Si-Si bonding is observed with higher annealing temperatures, as shown in Figure 2, suggesting a growth of the Si-ncs through the annealing process.

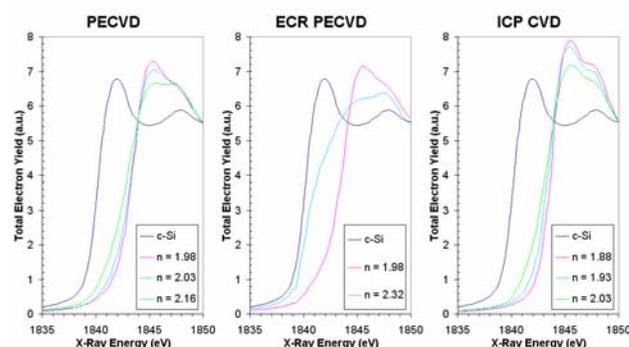


Figure 1: XAS spectra at the Si *K*-edge for as-deposited samples of varying composition fabricated by each deposition system compared with a reference crystalline silicon spectra. The absorption edge of samples in each system may be observed to push towards lower energies with increasing excess silicon concentration (larger *n*) corresponding to an increase in Si-Si bonding.

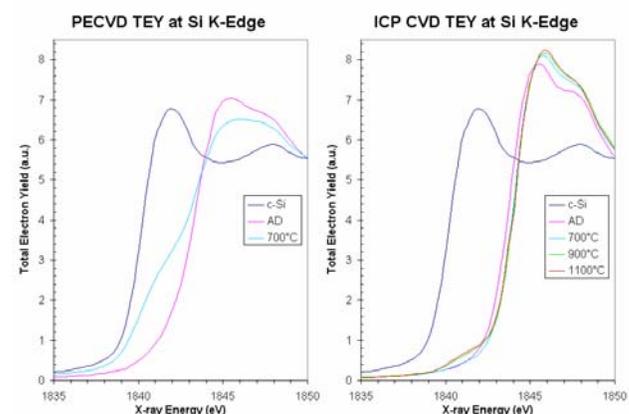


Figure 2: XAS spectra at the Si *K*-edge for PECVD ($n \approx 2.0$) and ICP CVD ($n \approx 1.9$) samples annealed for 60 minutes at different temperatures using a pure nitrogen ambient gas. For the PECVD sample, a large shoulder can be observed at the Si-Si bonding energy when annealed at 700°C. A similar shoulder begins to form in the ICP CVD sample at 900°C and grows larger when the annealing temperature is increased to 1100°C.

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