On the dynamics of phase separation under laser irradiation of Si/Ge rich oxides



ADVANCED RESEARCH LABORATORIES





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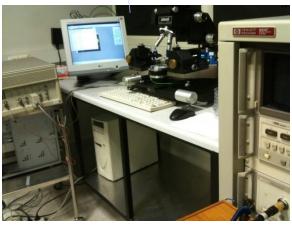




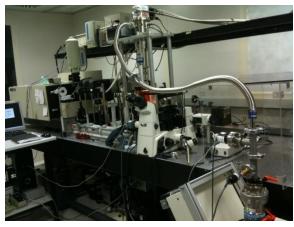


Advanced Research Laboratories (ARL)







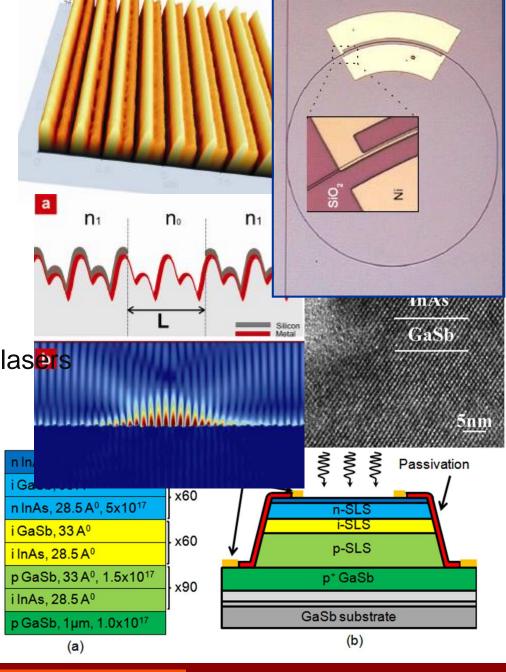






Research Portfolio

- InGaN/GaN Blue LEDs
- Type II SL IR photodetectors
- Quantum cascade lasers
- Si/Ge nanocrystals for solar cells
- Plasmonic cavities
- Plasmon-exciton interactions
- InGaAs/AlGaAs High power diode lasers
- Laser-matter interactions



Si/Ge nanocrystals for solar cells

Solar Cells: First Generation

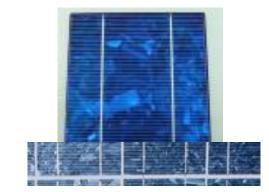
Single crystal Si

Multi crystal Si

Si single crystal based PV solar cells



Market share: % 35



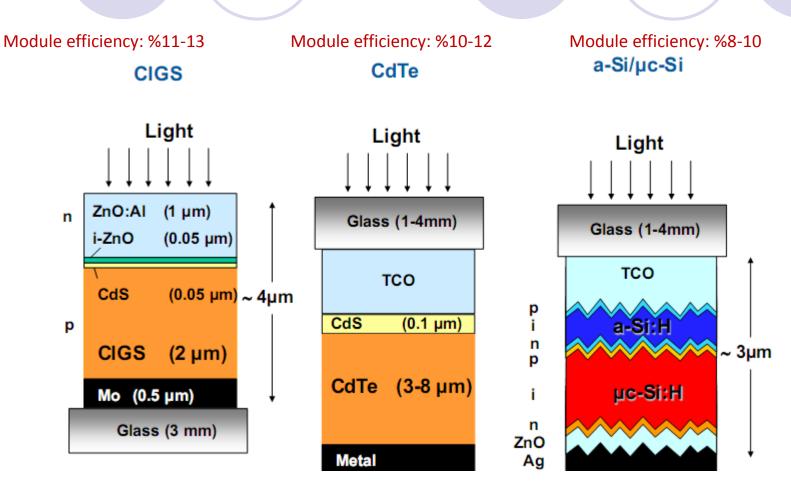
Module efficiency: %13-16



mc-Si PV Solar Module

Market share: % 49

Solar Cells: Second Generation



Market share <%1

Market share: %8

Market share: % 5

<u>THIN FILM SOLAR CELLS</u>

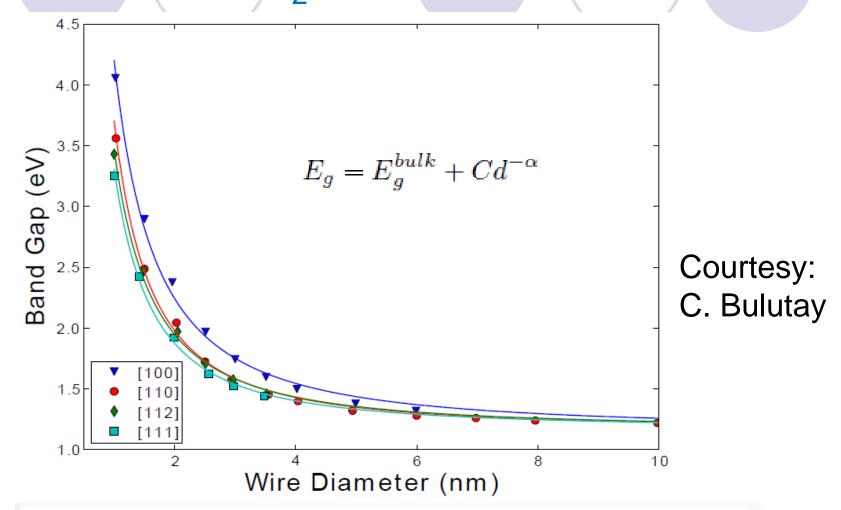
Third Generation Solar Cells

Any solar cell concept with limiting efficiency exceeding the single junction limit

3rd generation solutions
Nanosponge, Si/Ge Nanocrystal Networks

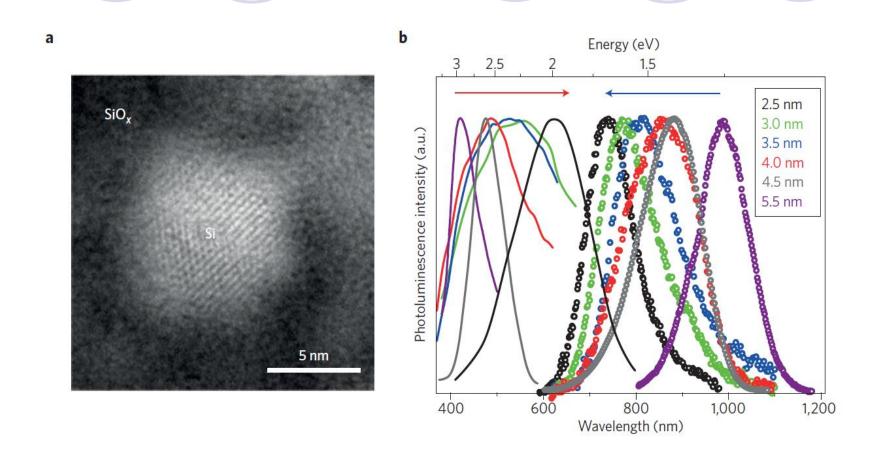
Main Motivation: lower cost, higher efficiency and/or lower production cost

Motivation: bandgap variation with the size Si Nanowires in SiO₂



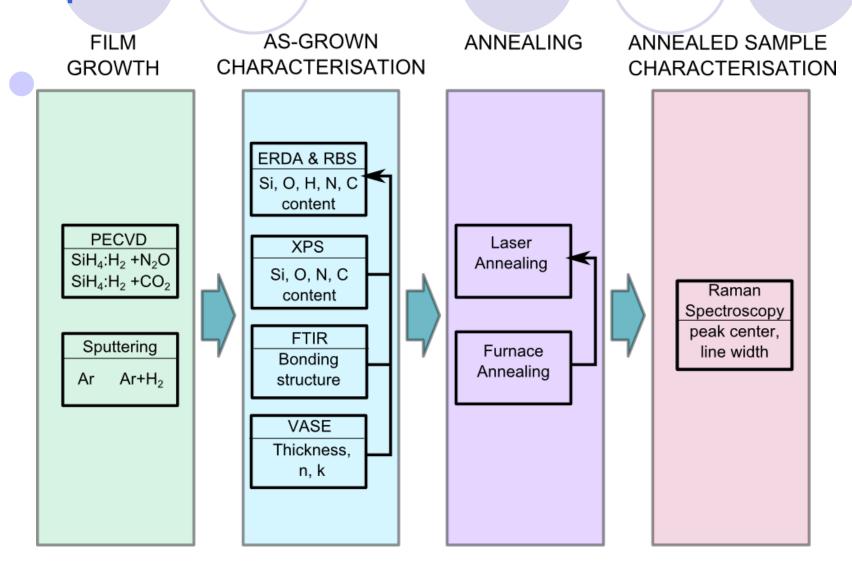
- Ma, D. D. D., C. S. Lee, F. C. K. Au, S. Y. Tong, and S. T. Lee, 2003, Science 299, 1874.
- [2] Yan, J.-A., L. Yang, and M. Y. Chou, 2007, Phys. Rev. B 76, 115319.
- Delerue, G. Allan, and M. Lannoo, 1993, Phys. Rev. B 48, 11024.

Bandgap variation with the size



Priolo et al., Nature Nanotechnology, 2004

Experiment





PECVD

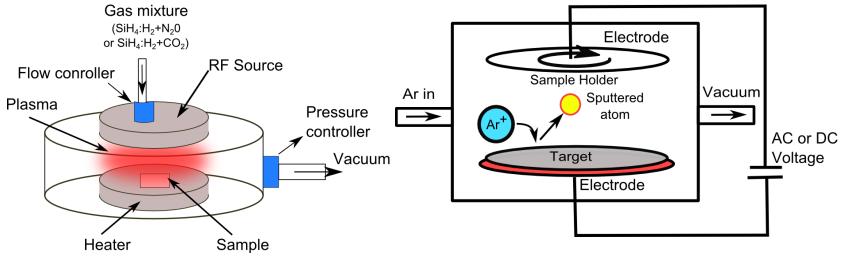
H series : SiH_4 : $H_2 + N_2O$

N series : $SiH_4:N_2 + N_2O$

C series : $SiH_4:H_2 + CO_2$

Sputtering

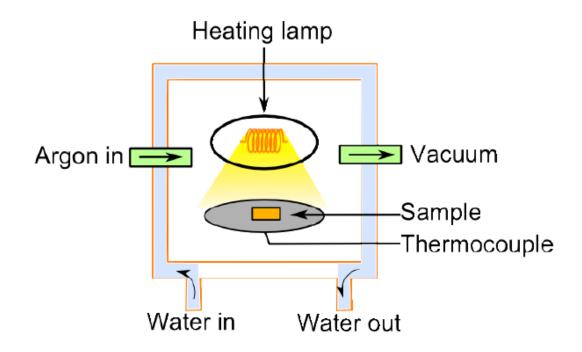
Base pressure	$4.5 \times 10^{-6} \text{ Torr}$
Working pressure	4 mBar
Process gases	Ar 20 sccm H ₂ 4 sccm
Process temperature	Room temperature
Time	1 hour
Power (Si)	54 W DC
Power (SiO ₂)	180 W RF
Film thickness	250 nm



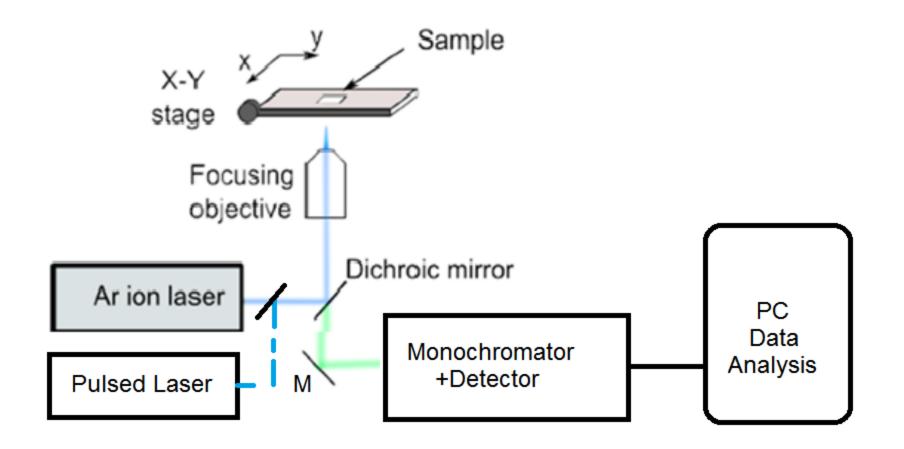
O/Si ≈1

Experiment: Furnace Annealing

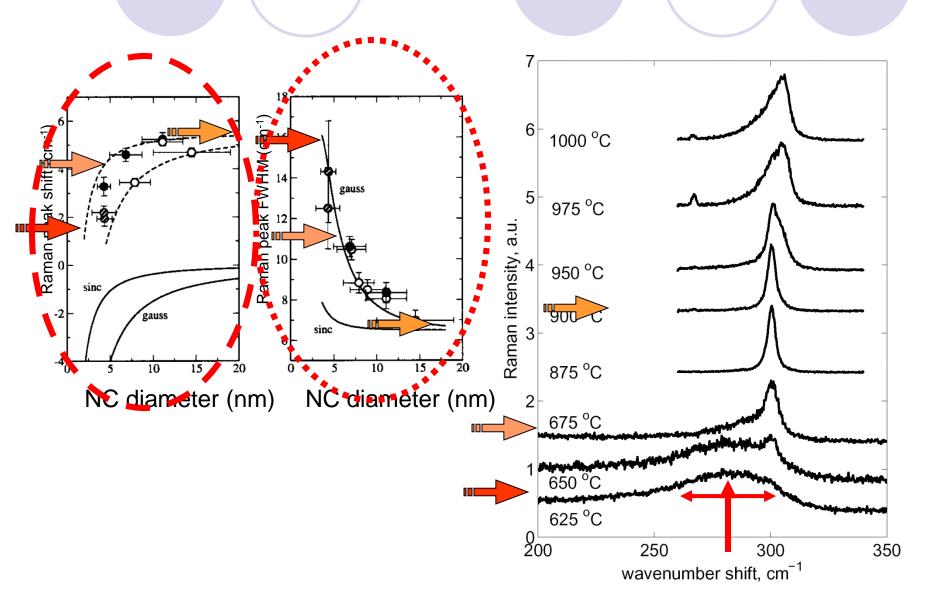
(Rapid Thermal Procesing)



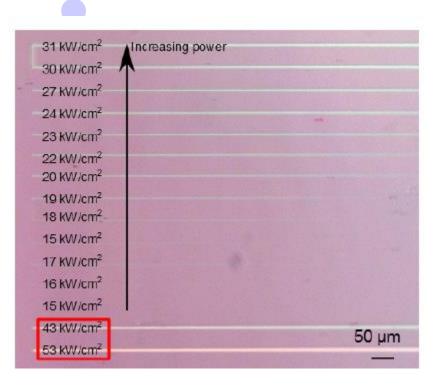
Experiment: Laser processing

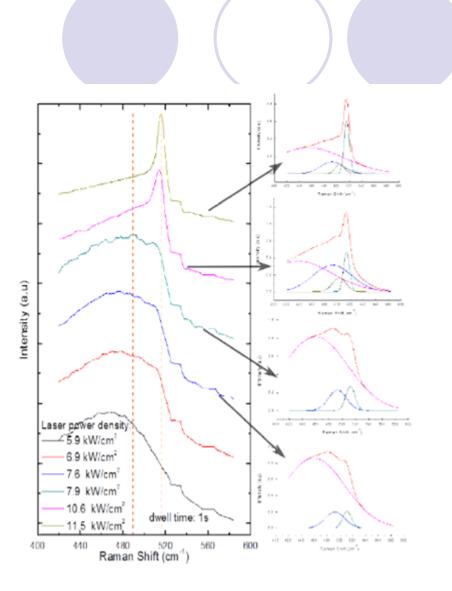


Raman characterization



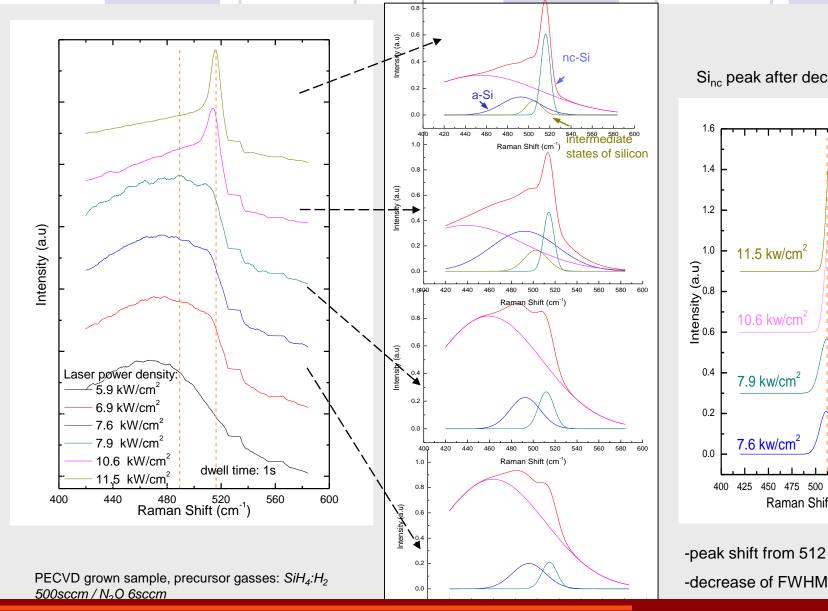
Laser Processing



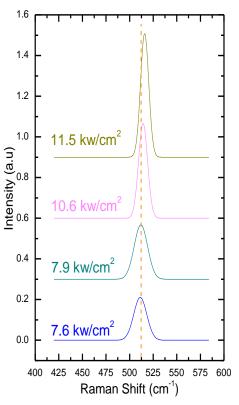


Sample H3

Raman spectroscopy analysis of laser annealed SiO_x films: effect of irradiation laser power density.



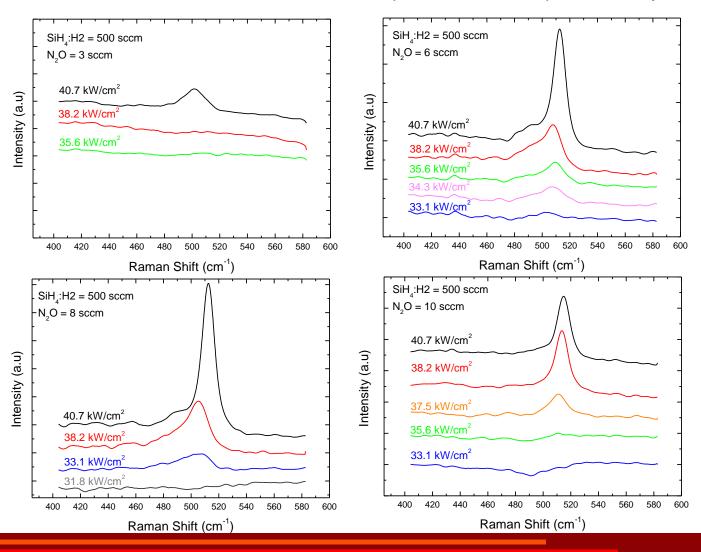
Sinc peak after deconvolution



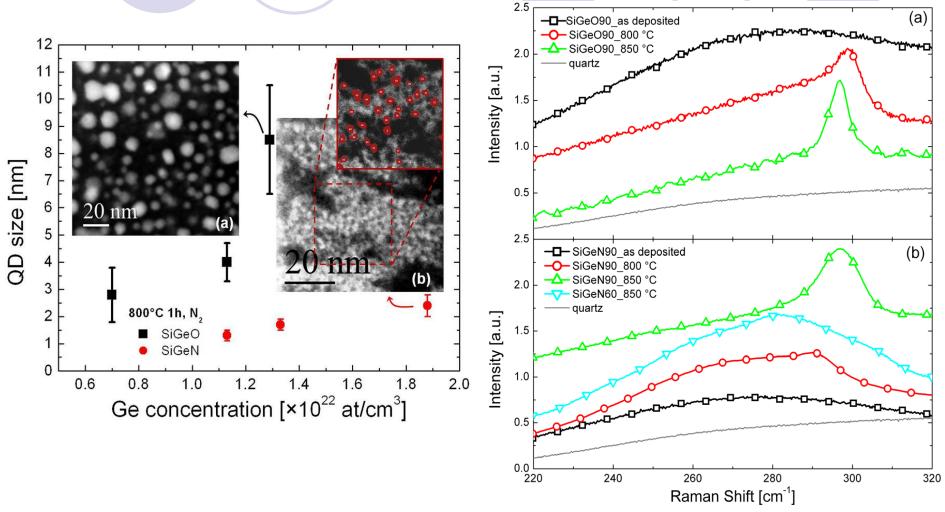
-peak shift from 512 to 516 cm⁻¹

Raman spectroscopy analysis of laser annealed SiO_x films: effect of composition.

Raman spectra of PECVD grown hydrogenated SiO_xN_y films with different precursor gas flow ratios, after irradiation with Ar+ laser beam up to 40.7 mW/cm² power density.



Ge Nanocrystals embedded in SiO₂ and Si₃N₄



Light harvesting with Ge quantum dots embedded in SiO₂ or Si₃N₄

Cosente et al., J.Appl. Phys., 115 043113, 2014

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The role of the interface in germanium quantum dots: when not only size matters for quantum confinement effects**

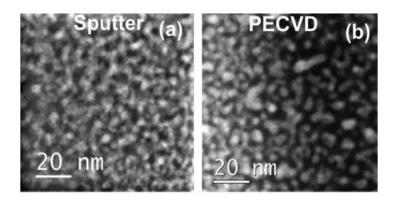
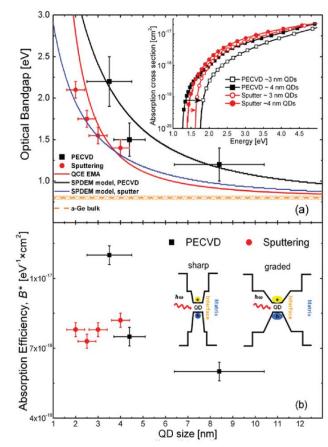


Fig. 1 Typical cross sectional HAADF STEM images of Ge QDs in SiO₂. Bright spots correspond to Ge QDs obtained by sputter (a) or PECVD techniques (b) from SiGeO films having $\sim 1.3 \times 10^{22}$ at cm⁻³ of Ge.



Ge nanocrystals in SiNy

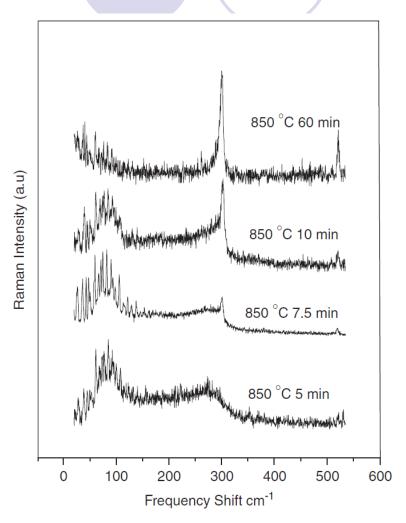


Fig. 2. Raman spectra of SiN_x :Ge films annealed in vacuum at 850 °C for various durations. Growth of Ge phonon mode at $300 \, \mathrm{cm}^{-1}$ as a function of annealing time observed, without any quenching.

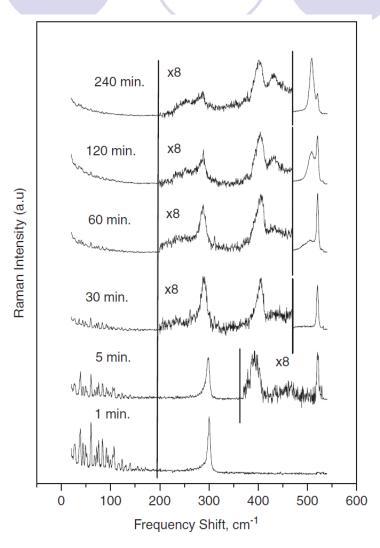
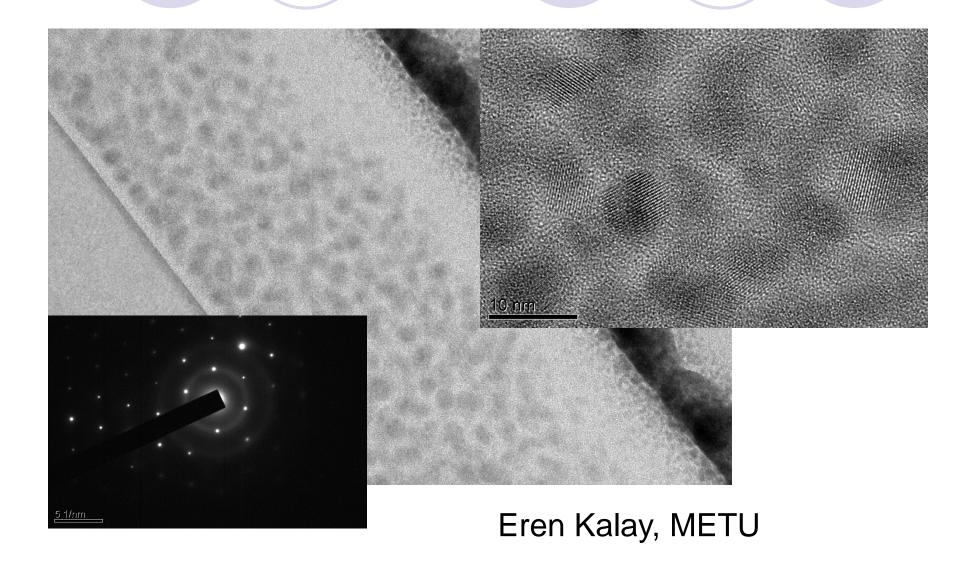


Fig. 3. Raman spectra of samples with prolonged vacuum anneal of ${\rm SiN}_x$: Ge films at 1050 °C.

Structural Analysis: TEM



Proposal

- Study in-situ dynamics of phase separation during cw/pulsed laser irradiation of Si/Ge rich oxides.
- Study in-situ crystallisation dynamics, using short pulse Xrays produced in PETRA III. Very fine collimated beams may make it possible to study extremely small nanocrystals of <10 nm.
- Grazing Incidence Nuclear Resonant Scattering for morphological and structural changes along with mapping of the strain on the nanocrystals, effecting their optical and electronic properties.

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Thank you!