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Harsh Gupta, Olivier Plantevin, Ravi Bommali, Santanu Ghosh, Pankaj Srivastava. A Study of Temperature-Dependent Photoluminescence from As-Deposited and Heavy-Ion-Irradiated Plasma-Enhanced Chemical Vapor Deposition-Grown Si-Rich  $a$  -SiN  $x$ :H Thin Films. *physica status solidi (b)*, 2020, 257 (5), pp.314001. 10.1002/pssb.201900378 . hal-04280241

**HAL Id: hal-04280241**

**<https://hal.science/hal-04280241>**

Submitted on 22 Nov 2023

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**Study of temperature dependent photoluminescence from as deposited and  
heavy ion irradiated PECVD grown Si rich a-SiN<sub>x</sub>:H thin films**

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Keywords: Si rich silicon nitride, temperature dependent photoluminescence, swift heavy ion irradiation, Activation energy

Temperature Dependent Photoluminescence (PL) measurements in the range of 10 K to 300 K were performed on silicon rich hydrogenated silicon nitride films deposited by plasma-enhanced chemical-vapor deposition. In-situ formation of Si QDs with average size of 3.5nm was observed. Composition of thin films was estimated by Rutherford back scattering and ellipsometry. A broad PL spectrum from thin films is decomposed into contributions from Si QDs, N dangling bonds and Si dangling bonds (K-centers). At low temperature a strong PL can be observed, its quenching with increase in temperature is explained by thermal activation of non-radiative decay mechanisms. Further, temperature dependent PL for swift heavy ion irradiated films show similar quenching with increase in temperature. However, the effect of irradiation on the luminescence mechanisms of *a*-SiN<sub>x</sub>:H is revealed at low temperatures (<120K) only, whereby a relatively narrow peak around 2.0 eV emerges. These results clearly show the role of radiative defects in the luminescence from Si rich Silicon nitride.

With the observation of visible light emission in porous Si, vigorous research activities focused on Si nanostructures were initiated for their potential application for the Si-based optical devices.<sup>[1]</sup> Silicon nanostructures show a quantum confinement effect, which results into an enhanced rate of electron–hole radiative recombination, luminescence and affects many other properties.<sup>[2,3]</sup> In recent years, an extensive research on silicon nanocrystals embedded in a dielectric matrix has been conducted because of their potential for applications in silicon-based optoelectronic devices, SiO<sub>2</sub> and Si<sub>3</sub>N<sub>4</sub> are two of the most suitable dielectric matrices to host Si quantum dots (QDs).<sup>[4–6]</sup> But a large band gap of the SiO<sub>2</sub> (~9eV) in comparison to Si<sub>3</sub>N<sub>4</sub> (~5eV) constraints the injection of the carriers, it reduces the efficiency and reliability of the devices, making nitride matrix a better candidate for electrical and optical devices.<sup>[7,8]</sup> Despite expected advantages from Si<sub>3</sub>N<sub>4</sub> as a matrix material for Si NCs, the fundamental origin of the luminescence from Si QDs in Si<sub>3</sub>N<sub>4</sub> system is still not completely understood. Many researchers have attributed the photoluminescence (PL) to quantum confined excitons in the nitride-embedded Si NCs or amorphous Si QDs.<sup>[2,9–11]</sup> Also, various radiative defects or band tail luminescence were suggested to explain the PL from silicon rich silicon nitride.<sup>[12–15]</sup> PL (color, intensity, stability) of silicon nanostructures embedded in silicon nitride are affected not only by the average size, size dispersion and density of Si nanostructures, but also by chemical composition of matrix.<sup>[15,16]</sup> Post-deposition treatments significantly changes the properties such as band gap, refractive index and size of Si QDs in a-SiN<sub>x</sub>:H thin films. Kachurin et al. have shown that swift heavy ion irradiation of 700 MeV results in formation of radiative recombination states in SiO<sub>2</sub> due to Si precipitation.<sup>[17]</sup> A shift and gradual increase in defect contribution in overall PL spectra at different excitations energies upon swift heavy ion irradiation at room temperature was reported by Bommali et al,<sup>[18]</sup> as out diffusion of hydrogen upon irradiation generate defects, which further modifies the PL from the thin films. Whereas, D. Murzalinov et al reported reversible quenching of visible PL upon

irradiation upon further annealing.<sup>[19]</sup> To the best of our knowledge temperature dependent PL study to understand the origin luminescence mechanism for as deposited and upon swift heavy ion irradiated a-SiN<sub>x</sub>:H thin films has not been reported so far.

In present work we report the temperature dependent PL from as-deposited and swift heavy ion irradiated hydrogenated silicon rich silicon nitride (*a*-SiN<sub>x</sub>:H) thin films with in-situ formation of Si QDs grown by plasma enhanced chemical vapor deposition (PECVD) to understand origin of radiative defect states and evolution of PL. A detailed study of PL quenching with temperature is performed in accordance with thermal activation of non-radiative decay mechanisms by the modelling of Arrhenius law.

Refractive index and thickness for as-deposited thin films as measured by single wavelength null ellipsometry are 2.1 and 280 nm respectively at 628 nm. Stoichiometry  $x$  ( $=[N]/[Si]$ ) calculated using measured refractive index value is found to be 0.56, though to calculate exact stoichiometry using ellipsometry is difficult as incorporated hydrogen also affects the optical properties of film but it clearly indicates that films are Si rich in nature as for stoichiometric films  $x= 1.33$ . Also,  $[N]/[Si]$  calculated by Rutherford Backscattering (**figure S1**) was found to be 0.58 which also establish the Si rich nature of as-deposited thin films. **Figure1** shows photoluminescence for as deposited Si rich a-SiN<sub>x</sub>:H thin films at different temperatures from 10 K to 300 K. We can observe a broad PL which quenches with temperature. Like other amorphous materials, a-SiN<sub>x</sub>:H also consists of a randomly-organized network of atoms. Bonding and compositional fluctuations introduce band-tail states observed around the edges, these band tail states can account for the broad PL spectra observed for a-SiN<sub>x</sub>:H. The observed PL data was deconvoluted and well fitted with three Gaussian peaks **figure 2**, indicating the presence of three different radiative transitions. As the band gap and position of different states in a-SiN<sub>x</sub>:H depend significantly on the stoichiometry

of films, peaks contributing to PL spectra are expected to be arising from intra band states and from Si inclusions.

The different radiative transitions contributing to luminescence from such partially phase separated Si rich a-SiN<sub>x</sub>:H thin films are distinct defect states transitions including N dangling bonds (DBs, ~2.04 eV), Si DBs (K<sup>0</sup> centers ~2.25 eV), and Si QDs (a-Si:H inclusions, ~1.84 eV).<sup>[9,14,20–25]</sup> A schematic of a-SiN<sub>x</sub>:H band diagram (figure 1) shows various intra-band radiative defect states. In **figure 3** transmission electron microscopy (TEM) image shows in-situ formation of Si QDs in Si rich a-SiN<sub>x</sub>:H film due to phase separation. The size distribution of QDs is shown in figure 3, average size of QDs is found to be ~3.5nm. Based on an effective mass theory assuming an infinite potential barrier, the energy gap  $E$  for three-dimensionally confined a-Si QDs can be expressed as equation 1: <sup>[3,10]</sup>

$$E(\text{eV})=E_{(\text{bulk})}+[C/d^2] \quad (1)$$

where  $E_{\text{bulk}}$  represents the bulk a-Si band gap(~1.6eV),  $d$  the dot size (nm) and  $C$  the confinement parameter (as reported in literature value of  $C$  to be 2.40).<sup>[3]</sup> Using above parameters and equation 1 with an average size ( $d$ ) as 3.5nm, the energy band gap in our case turns to be 1.79eV, which is in close agreement with our observed PL spectra peak corresponding to Si QDs (~1.84eV). At higher temperatures radiative transitions (mainly Si QDs, Si DBs) are being quenched predominantly by competing non-radiative transitions such as multi-phonon relaxation or phonon assisted energy transfer. As with an increase in temperature phonon assisted transitions become more and more significant, the energy is given up as heat to the lattice during the process. Therefore, with an increase in temperature the overall integrated PL intensity grows weaker and weaker.

Further, to understand better the quenching of PL with increase in temperature explained by thermal activation of non-radiative decay mechanisms, activation energies for different competing non-radiative recombination are calculated. To calculate the activation energy corresponding to the non-radiative recombination processes we fit experimental data of PL intensity as a function of temperature into single exponential Arrhenius equation (**figure S2**) equation 2. [26–28]

$$I(T) = \frac{I(o)}{1 + AT^{3/2}e^{-E_a/kT}} \quad (2)$$

Where  $I(o)$  is intensity at 10K,  $E_a$  activation energy for non-radiative decay mechanism, fitting parameter  $A$  and  $k$  Boltzmann constant. Also, in the model the  $T^{3/2}$  pre-factor depicts the temperature dependence of the effective density of states at the band edges. This temperature pre-factor before the exponential terms is used to indicate thermal depopulation of the bound states into the conduction/valence band edges. Fitting this equation by best fit parameters, we could estimate  $E_a$  for transitions centered around 1.84eV, 2.04V and 2.25eV respectively fitted with equation 2. All the activation energies calculated as per Arrhenius equation are summed up in **table 1**. For various non-radiative recombination such as multi-phonon relaxation or phonon assisted energy transfer calculated activation energies for all the transitions contributing towards PL are below thermal energy at room temperature ( $\sim 25$  meV), which explain quenching of PL from as-deposited thin film with increase in temperature, well below room temperature.

Transition(eV)	Si dbs (2.25eV)	N dbs (2.04eV)	Si QDs (1.84eV)
<b>Ea (meV)</b>	13.5	10.4	9.4

Table 1: Activation energy calculated using Arrhenius equation for different transitions.

**Figure 4** shows temperature dependent photoluminescence for Si rich a-SiNx:H thin film irradiated with swift heavy 100 MeV Au<sup>8+</sup> ions from 20 K to 300 K, at lower temperatures a-SiNx:H thin film shows strong PL which quenches rapidly with temperature. Similar trend was observed in as-

deposited thin film. Irradiated film shows more complex PL than as-deposited a-SiN<sub>x</sub>:H thin films. The distinct radiative transitions involved in luminescence from swift heavy ion irradiated Si rich a-SiN<sub>x</sub>:H arises from different radiative defect states such as N dangling bonds (DBs, ~2.04 eV), Si DBs (K<sup>0</sup> centers ~2.23 eV and ~2.32 eV), and Si QDs (a-Si:H inclusions, ~1.84 eV).<sup>[20,22,23]</sup>

In **figure 5** deconvolution and fitting of PL spectra using Gaussian peaks at different temperature clearly shows presence of additional radiative defect states in irradiated thin films. Swift heavy ion irradiation of a-SiN<sub>x</sub>:H thin film leads to out-diffusion of hydrogen from lattice, which comes from breaking of Si-H and N-H bonds.<sup>[29],30]</sup> Therefore irradiation generates a greater number of defect states (both N dangling bonds and Si dangling bonds) in a-SiN<sub>x</sub>:H, which contribute to overall PL from irradiated thin films. Also, there is a gradual shift in peak positions of different radiative recombination with increase in temperature. A schematic of a-SiN<sub>x</sub>:H band diagram (figure 4) shows intra-band radiative defect states. To understand the quenching of PL in irradiated thin films with an increase in temperature, activation energies for different competing non-radiative recombination was calculated by fitting experimental data of PL intensity as a function of temperature into single exponential Arrhenius equation (**figure S3**) equation 2. Similar to as-deposited thin films, calculated activation energies for all the transitions contributing towards PL for irradiated thin films are given in **table 2**, for all different thermal activation of non-radiative decay are below thermal energy at room temperature (~25 meV). Hence the non-radiative decay starts to dominate well below room temperature resulting in quenching of PL with temperature.

Transition (eV)	Si dbs (2.32eV)	Si dbs (2.18eV)	N dbs (2.05eV)	Si QDs (1.84eV)
<b>E<sub>a</sub> (meV)</b>	12.5	7.8	14.01	16.8

Table 2: Activation energy calculated by Arrhenius equation for different transitions.

When compared to as-deposited thin film, irradiated thin film show different PL emission. **Figure S4** shows that at low temperature, we can observe sharper contribution from additional defects states in case of an irradiated thin film. This sharp contribution to the PL clearly arises from the defects created during the irradiation process and indicates that the defect states are energetically less dispersed as compared to an as-deposited film. The narrow distribution of defects might have its origins in the specific energy loss of ion beam employed in these experiments. Further with an increase in temperature luminescence from the irradiation induced defects states quenches rapidly but overall contribution of defect state PL is still dominant in irradiated thin film at room temperature. At higher temperature we can clearly see more quenching of Si QDs and N dangling bonds contribution of PL from as-deposited than in irradiated thin film. Irradiated thin films shows significant contribution in PL from N dangling bonds and Si dangling bonds at room temperature as compared to as-deposited thin film due to generation of above-mentioned defect states. Also, generation of greater number of defect states is the reason behind increased activation energy of non-radiative decay recombination for different radiative states in case of irradiated film compared to as-deposited thin film.

In summary RBS and refractive index studies of thin films confirms that films are Si rich silicon nitride in nature with a stoichiometry of  $x=0.58$ . TEM image confirms the in-situ formation of Si QDs of average size of 3.5nm. We have shown a comparative study of temperature dependent PL emission from as-deposited and swift heavy ion irradiated thin films. For as-deposited film at low temperatures Si rich silicon nitride with in-situ Si QDs show strong PL at low temperatures, the overall PL is a contribution of three different radiative transitions. Also, with increase in temperature intensity of emission decreases drastically. This decrease in PL intensity with temperature is explained by thermal activation of non-radiative decay mechanisms, which suggest that activation energies for competing non-radiative recombination (7.4-13.5 meV) in process are



lesser than thermal energy at room temperature. Hence with increase in temperature rate of non-radiative recombination increases which results in weaker PL from a-SiNx:H films at room temperature. In case of irradiated thin films, temperature dependent PL shows presence of additional radiative defect states, which are generated as a result of out-diffusion of hydrogen upon swift heavy ion irradiation. PL from irradiated films also quenches with increase in temperature like as-deposited films. Due to increase in defect states over all activation energy for non-radiative recombination increases (8.89-16.8 meV) for irradiated films which is still lesser than room temperature thermal energy, explaining quenching of PL at room temperature.

### Experimental Section

A set of a-SiNx:H films was grown on n-Si by PECVD using SiH<sub>4</sub> (4% in Ar) and NH<sub>3</sub> as precursor gases at a pressure and temperature of 1 mbar and 200<sup>0</sup>C respectively. Film thickness and refractive index were calculated by single wavelength ellipsometry at 628 nm with incident angle of 70<sup>0</sup>. Film stoichiometry x (x=[N]/[Si]) was calculated using equation 3, where n is the refractive index of the as-deposited films, while  $n_{aSi:H}$ ,  $n_{aSi_3N_4}$  are refractive index of a-Si:H and Si<sub>3</sub>N<sub>4</sub> respectively.<sup>[31]</sup>

$$x = \frac{4(n_{aSi:H} - n)}{3(n + n_{aSi:H} - 2n_{aSi_3N_4})} \quad (3)$$

Rutherford backscattering (RBS) spectra was recorded for a-SiNx:H thin films using He<sup>+</sup> ions of energy 2 MeV. The temperature dependent PL (10 K to 300 K) was recorded for thin films using excitation wavelength of 488 nm. Swift heavy ion irradiation (Au<sup>8+</sup>) of energy 100MeV at room temperature with a fluence of 5x10<sup>12</sup> ions cm<sup>-2</sup> was performed by using a Pelletron Accelerator at the Inter University Accelerator Centre, New Delhi, India. At this energy, electronic energy losses ( $E_e \sim 931 \text{ eV } \text{\AA}^{-1}$ ) dominate over nuclear energy losses ( $E_n \sim 1.70 \text{ eV } \text{\AA}^{-1}$ ) as per the calculations from Stopping power and Ranges of Ions in Matter (SRIM).<sup>[32]</sup>

## Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

## Acknowledgment:

Author acknowledgement UGC, India and IIT Delhi for financial support and resources. IUAC, New Delhi is acknowledged for swift heavy ion irradiation facility.

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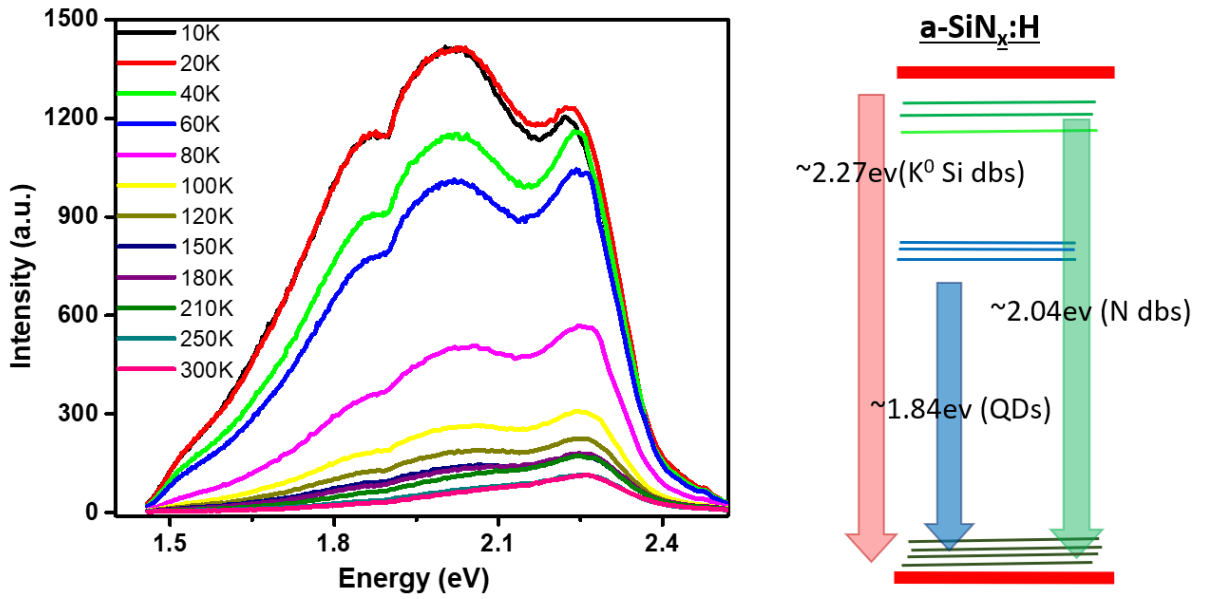
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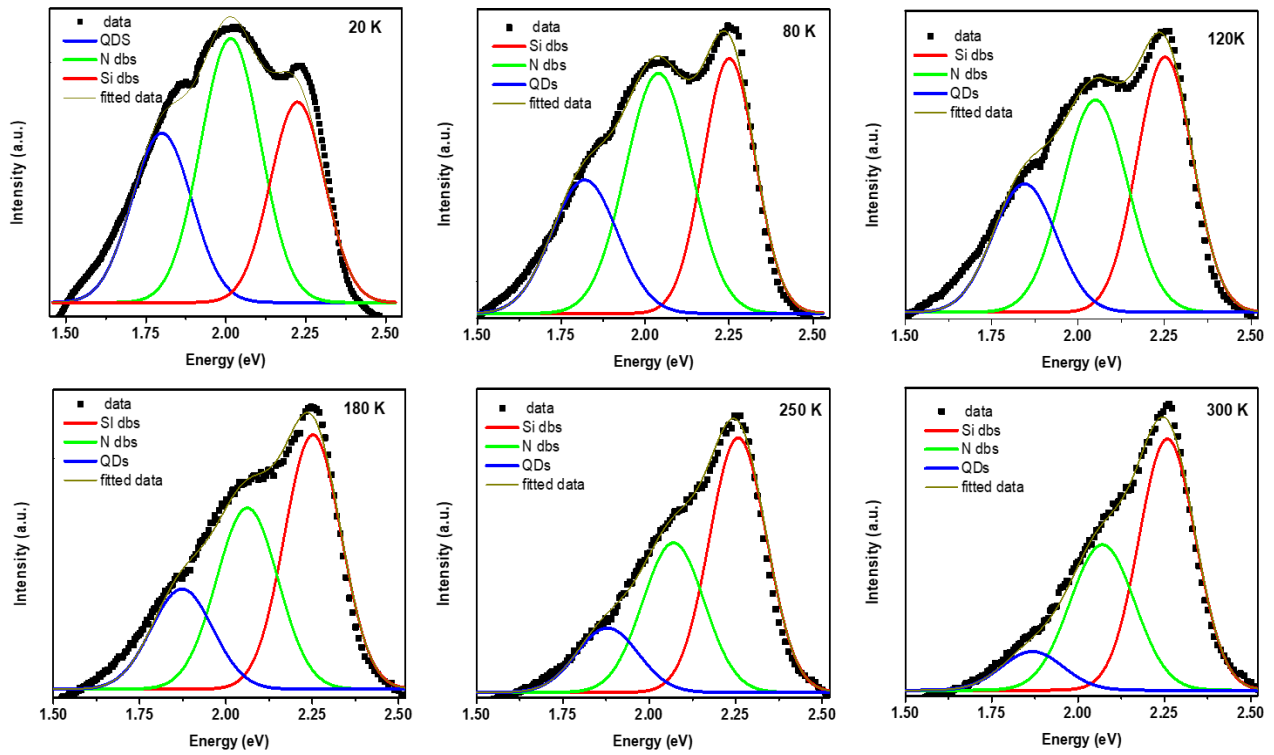
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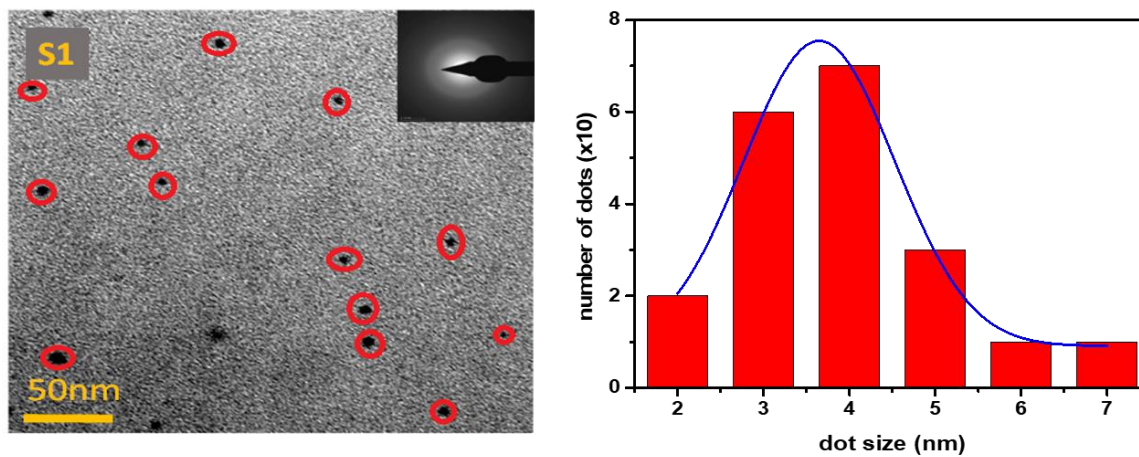
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**Figure 1:** Temperature dependent PL of as deposited a-Si<sub>x</sub>N<sub>y</sub>:H thin films and schematic diagram for radiative transitions

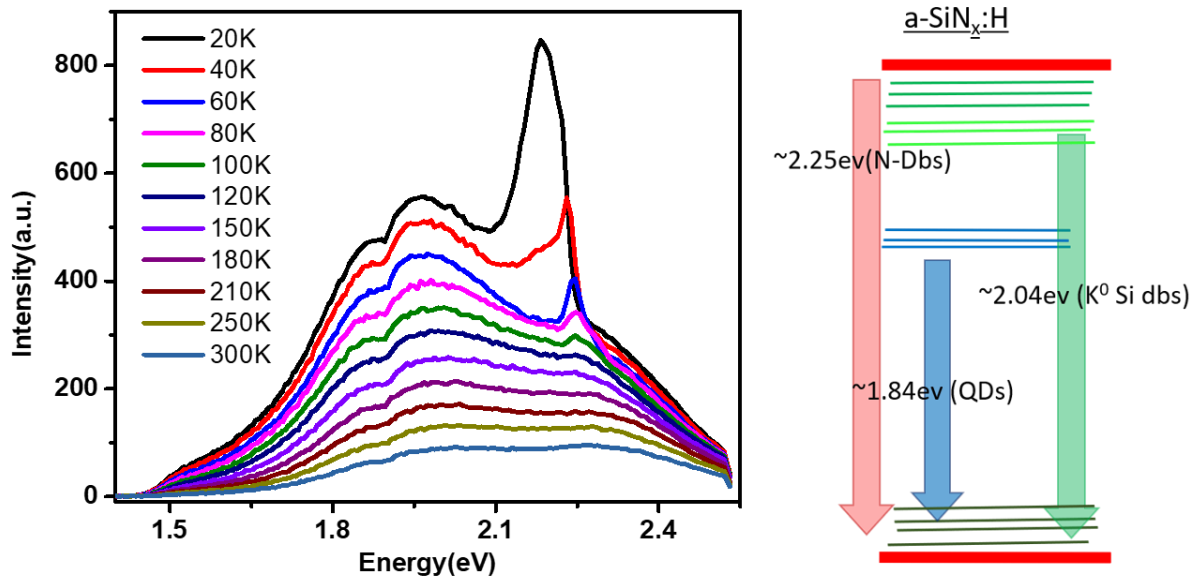


**Figure 2:** PL spectra of as-deposited a-SiNx:H at different temperature deconvoluted and fitted with Gaussian peaks.

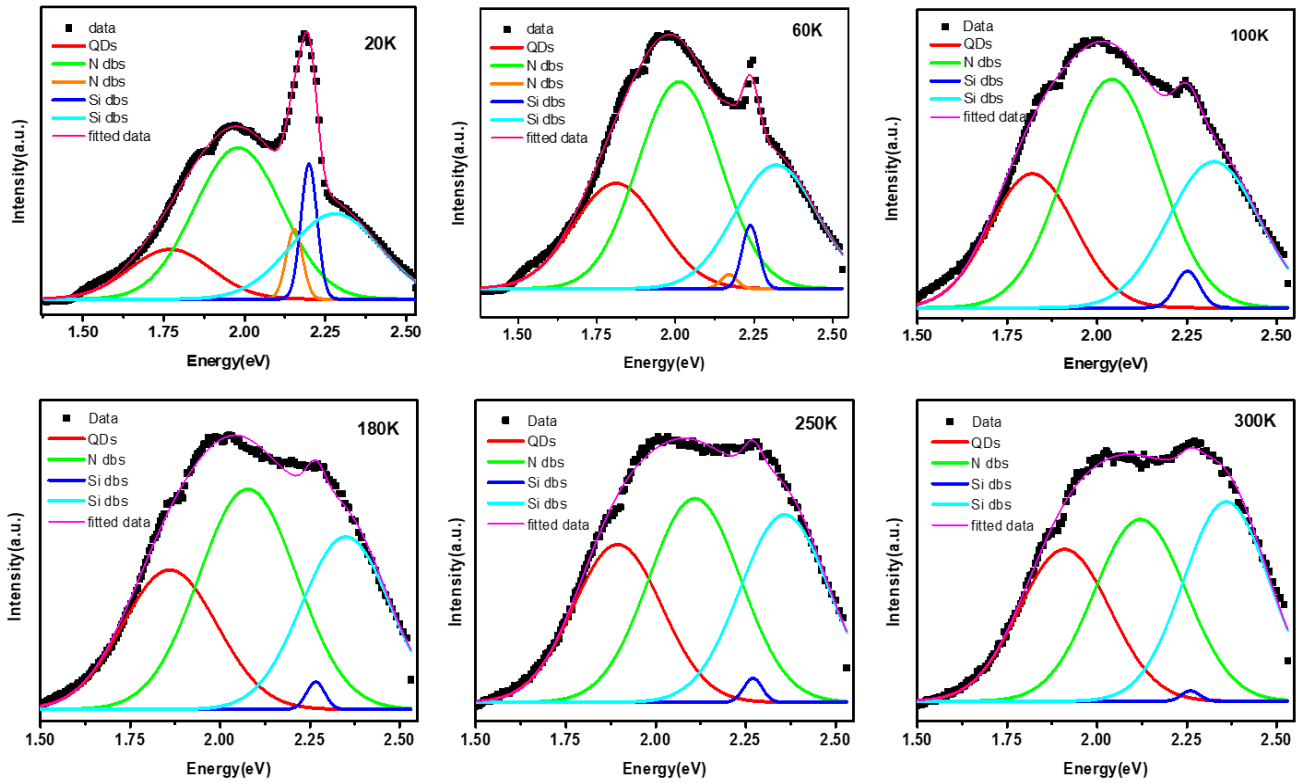


**Figure 3** TEM image for as-deposited a-SiN<sub>x</sub>:H thin films showing presence of Si QDs and Size distribution for Si QDs.





**Figure 4** Temperature dependent PL of irradiated a-SiN<sub>x</sub>:H thin films and schematic diagram for radiative transitions.



**Figure 5** PL spectra of irradiated a-SiNx:H thin film at different temperature deconvoluted and fitted with Gaussian peaks.

**Table1** Activation energy calculated using Arrhenius equation corresponding to different transitions for as deposited thin film.

Transition(eV)	Si dbs (2.25eV)	N dbs (2.04eV)	Si QDs (1.84eV)
<b>E<sub>a</sub> (meV)</b>	13.5	10.6	7.4

**Table 2** Activation energy calculated by Arrhenius equation corresponding to different transitions for irradiated thin film

Transition (eV)	Si dbs (2.32eV)	Si dbs (2.18eV)	N dbs ( 2.05eV)	Si QDs (1.84eV)
<b>E<sub>a</sub> (meV)</b>	14.56	8.89	14.01	16.8