Silicon nanocrytallines composite sol-gel rod for optical applications

M. Naziruddin Khan

Department of Physics, Faculty of Science Islamic University of Madinah-42351, Saudi Arabia Email: [mnkhan_phy@yahoo.com,](mailto:mnkhan_phy@yahoo.com) mnkhan@iu.edu.sa

Abstract

Nanocrystallines silicon (NCs Si) powders of heterogeneous sizes were uniformly dispersed in some solvents. Different quantity of NCs Si powder $(0.00150/0.0012/0.0070)$ g was taken in the 10 ml of solvents to identify the effect of photoluminescence property. The NCs Si solution was directly incorporated in sol-gel matrix and developed nanocomposite rods. Light emitting from NCs Si solutions and nanocomposite rods were detected by exposing UV light. Optical properties of the NCs Si solution and nanocomposite rod were systematically studied using different characterization techniques. The effect on the properties of NCs Si in the environment of sol-gel matrix was studied. The significant change in absorption and emission property of NCs Si solution is observed when embedded in sol-gel matrix. The predicted band gap 2.8 eV of NCs Si in solution was changed to 3.63 eV when it is included in solid matrix. Crystalline structures of different sizes of NCs Si are identified by XRD and TEM. Well presence and distribution of NCs Si in the sol-gel surface were revealed by SEM images and EDX spectra. Spontaneous emissions (SE) of NCs Si solution and nanocomposite sol-gel rods were tested using third harmonic 355 nm laser source of a pico second tunable laser system. The enhanced SE signal from the nanocomposite solid rod is quite significant, which indicates that SE signal may enable to increase if highly packed NCs Si colloidal solution employ in sol-gel rod. The fabrication of light emitting composite solid rod may use as a solid state active medium for the ASE testing in the future work.

Keywords: Nanocrystals silicon, Sol-gel, Absorption, Emission, Spontaneous Emission and pico-second laser source

قضبان هالم السيليكون النانوية المركبة للتطبيقات البصرية

الملخص: تم تفريق مساحيق السيليكون النانوية) (Si NCsذات األحجام غير المتجانسة بشكل موحد في بعض المذيبات. تم أخذ كمية مختلفة من مسحوق NCs (0.0070 / 0.0012 / 0.00150)جم في 10 مل من المذيبات لتحديد تأثير خاصية التأللؤ الضوئي. تم دمج محلول Si NCs مباشرة في مصفوفة gel-sol وتطوير قضبان مر ّكبة نانوية. تم الكشف عن الضوء المنبعث من محاليل NC وقضبان المركبات النانوية عن طريق تعريض ضوء األشعة فوق البنفسجية. تمت دراسة الخواص البصرية لمحاليل Si NCs وقضبان المركبات النانوية بشكل منهجي باستخدام تقنيات توصيف مختلفة. تمت دراسة التأثير على خصائص Si NCs في بيئة مصفوفة gel-sol. لوحظ التغيير الكبير في خاصية االمتصاص واالنبعاث لمحلول Si NCs عند تضمينه في مصفوفة gel-sol. تم تغيير فجوة النطاق المتوقعة 2.8 فولت من Si NCs في المحلول إلى 3.63 فولت عندما يتم تضمينها في مصفوفة صلبة. يتم تحديد الهياكل البلورية ذات األحجام المختلفة من Si NCs بواسطة XRD و TEM. تم تأكيد وجود جيد وتوزيع Si NCs في سطح gel-sol بواسطة صور SEM وأطياف EDX. تم فحص االنبعاثات العفوية) (SEلمحلول Si NCs وقضبان gel-sol المتناهية الصغر باستخدام مصدر ليزر 355 نانومتر التوافقي الثالث لنظام ليزر بيكو قابل للضبط الثاني عالي الطاقة. تعد إشارة SE المح ّسنة من قضيب صلب المركب النانوي مهمًا جدًا ، مما يشير إلى أنه قد يكون من الممكن تحسين إشارة SE في المستقبل إذا تم استخدام محلول Si NCs الغرواني المعبأ للغاية في قضبان -sol. gel قد يستخدم تصنيع القضيب الصلب المركب الباعث للضوء كوسيط نشط للحالة الصلبة الختبار ASE في العمل المستقبلي.

1. Introduction

In the past decades, silicon-based nanocrystallines (NCs-Si) /nanoporous (Psi) have been interested for new type of photoelectronic and informational materials. Extensively, bulk silicon has been treated as an incompatible material for optical applications because of its incapable to emit light and indirect band gap in nature [1]. After the detection of luminescent light from nanoporous (Psi) and nanocrystals silicon (NCs-Si) by Canham in 1990 [2], it changed the view on this material for optical devices. For Si integrated circuits [3], the researchers have activated especially in the preparation and characterization the light emitting of Psi and NCs-Si [4]. The emission property of silicon nanoparticles are multi-colors with size dependent, which are considered as potential materials for fluorescent tags, biological imaging and bioanalysis [5-8]. Interestingly, luminescence Psi and NCs-Si are brighter with significant stable to photo bleaching [9], tunable at multi wavelengths than florescent organic dyes [10]. Nanocrystals silicon has nontoxic behavior and attracted for the application in pollution [11].

In fact, the proper use of NCs-Si/Psi in the light emitting applications such as (LEDs or injection lasers) is still interested including for fundamental research as well as for waveguides, photodetectors, solar cells, gas sensors [12 - 14] etc. The main challenge is to achieve ASE or lasing in silicon for optical circuits because the basic component is the laser light source. For the goal, a proper study on the photoluminescence properties of NCs-Si or Psi solutions under different conditions are still required. The good stability of porous silicon with high concentration in solution still is one of the important issues [15]. In this regard, the deposition of NCs Si in an insulator matrix environment, such as SiO2 matrix [16] were performed by physical techniques [17-18] because it can control the influences of the interface between the NCs Si and matrix. In addition, to realize the PL efficiency, fabrication and crystallization of nanocrystal silicon in SiO2 at low percent was performed using high thermal annealing techniques [19, 20]. Efforts have been ensued to increase the emission property of NCs-Si in SiO2 using high thermal annealing process [21– 27]. However, the disadvantage of thermal annealing process is considered as incompatible for the addition of LEDs in electronic systems. The higher band gap of the nanocrystals silicon in SiO2 matrix leads to a drawback when it operates with voltage of LEDs [28]. Therefore, strong emission from silicon nanoparticles with high density and small band gap for the optimization of optoelectronic properties was suggested [29]. To stabilize and maintain the photoluminescence (PL) property in matrix, direct incorporation of silicon nanoparticles in different matrices such as sol-gel derived fine powder [30], solgel derived-films [31], composites block silica matrix [32], silica aerogels pellets composites [33], blocks of aerogel [34] and NCs Si in aerogels film [35] were performed.

 Indeed, inclusion of silicon nanoparticles in sol-gel matrix is easily controllable and able to combine with high density of nanopart icles in view of desired applications. Optical stability of silicon nanoparticles within the matrix at low temperature may lead to light stimulation, for this, our group have carried out a series of studies on the optical and structural properties of porous silicon in sol gel [36-38], ormosils [39] and polymer matrix [40). In previous, we used to dope the chemically synthesized porous silicon in the matrices which observed significant unstable of particles during the drying of the matrix except the polymer matrix. Preparation of high density colloidal porous silicon based solution by chemical etching route is a major issue [15]. On other hand, we doped commercially obtain nanocrystal silicon powder in sol-gel hosts [41-42]. Results revealed that observed optical property of silicon nanocrystalline powder was significantly stable as compared to porous silicon in sol gel matrix during aging. In the present study, commercially obtained nanocrystalines Si powder were dispersed in some solvents and prepared colloidal solutions. Emission light from NCs Si solutions was tested under UV lamp before use. The light emitting solution was doped in sol-gel matrix and investigated by different characterization techniques. Spontaneous emissions of solution and composite samples were examined using a laser source of pico second laser system. The obtained properties of silicon monocrystalline composite sol gel are systematically discussed.

2. Materials and Preparation method

The nanocrysts silicon powder was procured from M K Impex, Canada (98+% -pure), heterogeneous size range (APS: 5-25nm). The NCs powder was prepared as a colloidal solution. For colloidal solution, the different approximate concentration of NCs Si powder was taken and dispersed in known volume of different solvents, such as ethanol, methanol, ethyl acetate, DMSO, acetone, DMF, acetonitrile, and formamide. The solutions were ultra-sonicated for well dispersion and centrifuged to remove the residue particles. Then the solutions were exposed to an UV lamp (Cole-Parmer, USA) of 365 nm to detect the light emitting.

 The NCs Si composite material was prepared using sol-gel technique. In the process, the inorganic precursor TEOS (Si $(OC₂H₅)₄$) (Aldrich, 98%) was taken as the starting material and reacted with ethanol (Riedel-deHaen) for hydrolysis and polycondensation to make sols. In composition, about 15-20 ml of TEOS was mixed with 20-25 ml of ethanol and stirred about 25-30 min. Then, about 10 ml of formamide as DCCA was included into the solution. The mixed solution was continously stirred for another 15 min. After dropping of 1 ml nitric acid as catalyst in 15 ml of distilled water, it poured drop wise in solution and stirred about 10-15 minutes. The NCs Si of 0.00150 g concentration based formamide solution was directly added in the final sol. The final composite sol was separated in some polystyrene tubes and cuvettes for drying at room temperature. Before aging, composite sol in tubes and quartz was sonicated to insure a homogenous distribution of particles. The nanocomposite sol becomes a gel in the tube and cuvette cell after a few weeks. The gel samples were transformed into solid state rod in the polystyrene tube after 4-5 weeks. The composite containing tube was preserved at 55° C in an oven for few days to vaporize the liquid. The final product rod was performed cutting and hand polishing for PL and SE test by laser action.

3. Characterization

The optical, structural, and SE properties of nanocrystals silicon in solvents and sol gel were investigated. The different properties of the NCs Si samples were recorded at room temperature.

X-ray diffraction (XRD) spectra of the NCs Si powder were scanned using a PANalytical X'Pert X-ray diffractometer. Absorption and

emission were recorded in UV-visible-NIR spectrophotometer (Jasco 670) and Fluorescence Spectrophometer (Lumina Thermo) respectively. The morphological structure of the NCs Si in solvent and in sol-gel matrix was examined by a field emission scanning electron microscope (FESEM, JEOL, JSM-6380LA). Particle size of NCs Si in the solution and matrix at high resolution were determined by transmission electron microscopy (JEOL, JEM2100F). The spontaneous emission spectra were examined using laser source of Pico Second Tunable Laser System (Lotti III) and monitored with ICCD Spectrograph (Andor).

4. Results and discussion

The crystalline structure of NCs-Si powder was inspected by XRD. The XRD data shows the original pattern of NCs-Si powder. The pattern of the NCs Si is found to be a good correspond to peak position and relative intensity of the standard pattern, indicating the high purity of Si powder. The observed peaks at 28.41, 47.26, and 56.06 correspond to the crystal plane (111), (220), and (311), respectively, as shown in Figure 1.

Figure 1: X-Ray diffraction pattern of NCs Si powder

Different concentration of the NCs silicon powder was used in various solvents. The solvents were selected as some polar and nonpolar which are ethanol, methanol, ethyl acetate, acetonitrile, Acetone, formamide, DMSO, DMF, THF, dioxane. Some NCs Si solutions were detected the emission light under the exposure of UV lamp. The NCs Si containing formamide, DMF, and DMSO solutions were found to be good luminescent under the naked eye of UV lamp. No luminescent light was detected from the NCs Si in THF, dioxane, ethanol, methanol, ethyl acetate, and acetonitrile acetone, which are not discussed in this work. The luminescent and non-luminescent light from NCs Si in different solvents is listed in Table 1. In addition, the NCs dispersed ethanol, methanol, ethyl acetate, acetonitrile, acetone, THF, and dioxane solution were aggregated or precipitated. The digital image of luminescent NCs Si in solvents is shown in Figure 2.

Figure 2: The digital image of light emitting NCs Si in: (a) Formamide, (b) DMF, (c) DMSO solvent under UV exposure Table 1: Details of NCs silicon in different solvents

The light emitting from NCs silicon in some solvents is detected under the exposure of UV lamp. Thus, the formamide based solution was selected to study the different properties of the NCs Si using different characterization techniques. The property of light emitting NCs Si dispersed DMSO, and DMF solution is not detailed except the images as shown in Figure 2. The NCs Si dispersed formamide solution was then incorporated in sol-gel matrix and developed into nanocomposite rod in order to examine the stimulated emission spectra under tunable laser system. The crack free composite rods were successfully prepared. It was easily able to cut and hand polished without any harmful to dopant. After cutting and polished, the nanocomposite rod was used to investigate the different properties. The digital image of the light emitting polished rod with and without UV exposure is seen in Figure 3 (c, d). The light emitting nanocomposite rods with the evolution of sol to solid rod under UV exposure is displayed in Figure 3 (a). The emitting color indicates the well dispersal and existence of NCs Si in the matrix environment.

Figure 3: The digital image of NCs Si doped (a) sol (b) rods before cutting (c) rod after cutting and (d) rods under UV light exposure.

As clearly seen in figure 3 (b), the digital image of light emitting nanocomposite rod which is comparable to the solution one. Brightness of nanocomposite rods indicates the significant stable of the NCs Si in sol-gel solid environment.

4.1. Morphology and Size

Morphological structure of NCs Si in solvent and composite matrix was inspected by SEM at high magnification scale is shown in Figure 4 (i). The native of nanocrystallines silicon in solvent is quite identical and closely gathered in the surface and similar to sandy surface. When NCs Si in the dried sol-gel surface, the nanoparticles become mono distribution in the microamorphous matrix environment as seen in Figure 4 (ii). The structure of NCs Si in the dried sol gel indicates the particles lie in the homogenous amorphous surface of matrix. Additionally, the role of DCCA in sol-gel composition can control stressing in sol-gel surface during drying and aging. Involvement of formamide as DCCA in matrix may help to form crack free structure. This may reason to the well distribution of NCs Si and stable provide good formidable rods. In general, dopant particles are sometimes evaporated during the transition phase of sol gel drying process, but the use of DCCA in the process may slow down the evaporation of NCs Si. Therefore, NCs Si doped sol-gel rod were found to be more stable than the porous silicon (Psi) doped sol-gel rod [15, 38-39].

Figure 4. The SEM images of NCs Si in (i) solution and (ii) sol-gel matrix

Figure 5 (i) shows the typical TEM images for NCs Si solution at low and high resolution. Distribution of mono dark spot Si particles in the solvent is observed. Crystalline lattice structure of NCs Si particles about 3-7 nm size were detected as shown 5 (i-inset). Majority of Si particles are spherical with diameters ranging from 5 nm to 8 nm with highly crystalline in nature. NCs Si are distributed in sol-gel matrix with a roughly spherical shape as seen in Figure 5 (ii). The sizes of NCs Si are larger due to the envelope of sol-gel surface. Well presence of NCs Si in the solution is detected by EDX spectra as seen in Figure 5 (iii). The existence of NCs Si in the matrix is confirmed by EDX spectra as shown in Figure 5 (iv). Significant presence of NCs Si particles inside the surface of the matrix environment is witnessed by the image and EDX spectra which lead to enhance substantial strong emission light under UV light as shown in Figure 3.

Figure 5. The TEM images of NCs Si in (i) solution at low and high resolution (inset) (ii) sol-gel matrix (iii) EDX of solution and (iv) EDX of nanocomposite **4.2. Absorption property**

The absorption spectra of NCs Si in formamide at different concentration was measured. The absorbance is exponentially increased when the concentration increases in solution. There is no effect of the relative peak position relative to the concentration of NCs. The absorption spectra of NCs Si colloidal solution at different concentration are compared as shown in Figure 6 (i). Absorption spectra of NCs Si composite sol-gel matrix was recorded in the same spectral region. The absorbance and peaks of NCs Si in sol-gel are changed with broadening bandwidth. Shifting of peak and broadening the relative bandwidth may attribute to the solid environment of the matrix as listed data in Table 2.

Relative absorption peaks at 290 nm and 356 nm are observed in solution. The corresponding absorption peak of NCs Si are slightly shifted in sol-gel matrix as shown in Figure 6 (ii), the peaks 'a' and 'b'. The absorbance of the two peaks slightly increased in the sol gel. The comparison peaks of NCs Si in sol gel and solution are shown in Figure 6 (ii), a, b, c, and d peaks respectively.

Figure 6. Absorption spectra of NCs Si (i) solution at different concentration (ii) comparison with composite sol-gel matrix

4.2. Energy band gap

To examine the energy band gap of the NCs Si, the coefficient (α) was deduced from the absorption spectra using the relation

$$
\alpha=2.303\;\frac{A}{d}
$$

Where the parameter d and A are the thickness and the optical absorbance of the film.

The band gap of the NCs Si in solution and sol gel was calculated from Tauc's plots $[43]$ by explaining the relationship between coefficient (α) and photon energy for indirect band transition

$$
(\alpha h\nu) = C(h\nu - E_g)^n
$$

Where the parameters hv, C and E_g are the photon energy, a constant and the optical band gap respectively. The 'n' is a dependence parameter on the band of direct or indirect transition and the property of electron density in the valence and conduction bands.

The parameter $(ahv)^{1/n}$ was plotted with photon energy for different values of n. But, for the direct allowed transition, best fit was found when $n = 2$, whereas n=1/2 for an indirectly allowed transition. Such result indicates that the indirect transition behavior of noncrystalline materials. From the Tauc's equation for the allowed nondirect transition, the E_g ^{opt} (optical band gap) values can be obtained from the intercept of $(ahv)^{1/2}$ vs (hv) when $(ahv)^{1/2} = 0$.

The calculated band gap of NCs Si in formamide is shown in Figure 7(i). However, the band gap slightly changes from 2.9 eV of solution to 3.62eV in sol gel as displayed in Figure 7 (ii). The change in bandgap of NCs Si is due to the solid environment of matrix. The values of band gap for each NCs Si sample is detailed in **Table. 2.** Therefore, the optical band gap of NCs Si may depend on the physical environment of matrix and size.

Figure 7. Optical band of NCs Si in (i) formamide (ii) sol-gel matrix

4.3. Photoluminescence and spontaneous emission property

From the points of application, the photoluminescence (PL) mechanism of silicon nanoparticles in solid media was recorded and compared with the spectra of NCs Si solution. The excitation wavelength was selected as that exhibited the intense emission spectra and within the range of the absorption spectra of the NCs Si solution. The selected excitation wavelength was taken as an excitation pump laser source. For emission spectra, the sample was measured at different excitation wavelength to examine the intense emission peaks. Figure. 8 shows the emission and excitation spectra of NCs Si in formamide solvent and sol-gel matrix. It shows that the intense emission intensity was not observed at the excitation wavelength correspond to the strong absorbance peak of the NCs Si.

Significantly influence on the emission and excitation intensity is observed due to different concentration of NCs Si in solution. Higher concentration of NCs Si in solution is pronounced the stronger intensity as appeared at the emission peaks (a₁, b₁-442 nm) and excitation peaks (a₂, b₂-365 nm) as shown in Figure 8 (i). No major effect with peak position upon the variation of concentration is observed. The intensity of emission and excitation peaks of NCs Si solution are influenced when it included in the matrix environment. The emission and excitation spectra of NCs Si in solid sol-gel were recorded using different excitation wavelengths 365 nm, 355 nm, and 315 nm as shown in Figure 8(ii). The emission peak is substantially stronger at excitation of 365 nm than the excitation of 355 nm and 315 nm. For instance, the peaks a_1 , b_1 , and c are the emission of NCs Si in the sol gel at excitation of 365 nm, 355 nm, and 315 nm respectively as displayed in Figure 8 (ii). The excitation peak position of different solution observed at around 365 nm (a₂ and b₂) do not change as shown Figure 8 (i) and listed in Table 2. It means that stability of NCs Si in solgel environment during drying is much better than the porous silicon in sol-gel as our previously reported [36-37]. The emission intensity of NCs Si in sol gel matrix is quite significant as compared to intensity of NCs Si solution which may attribute to influence of temperature of sol gel $SiO₂$ environment during aging since the improvement in PL intensity of NCs Si in $SiO₂$ upon annealing temperature was observed [44]

The spontaneous emission (SE) from the NCs Si composite sol gel rod was examined by employing a 355nm pico second laser source. But intense emission of NCs Si in sol gel rod was attained at excitation of 365 nm as displayed in 8 (ii). Observation of NCs Si solution and composite sol-gel rod were conducted at the same condition under a high power tunable pico second laser source. The SE spectra of NCs Si solution were recorded at two concentrations. The NCs Si solution and composite rod were pumped at 3 mj, 6 mj and 10 mj laser energy as shown in Figure 9 (i-ii). There is exponentially increased in relative intensity with respect to concentration and pump energy. For instance, the observed SE intensity at 3 mj is weaker than the relative intensity of 6 mj as seen in Figure 9 (iii). It indicates that the luminescent intensity of the NCs Si depends on the pump energy. When the excitation energy increased at 10 mj, SE intensity of NCs Si solution is improved as shown in Figure 9 (iv). The behavior of SE intensity for NCs Si solution depends on the proper pump energy with the concentration. The SE stability of NCs Si in formamide is quite significant and more superior than the porous silicon in THF and dioxane under laser action [38-39].

Figure 9. Spontaneous emission spectra of NCs Si in (i-iv) solution at different concentration and pumped energy

Therefore, influence on spontaneous intensity of NCs Si solution is the factor of concentration, excitation wavelength, and pump energy. As clearly indicate that the exhibited light from NCs Si solution is about 5 times intense when the pump intensity increases from 3 mj to 6 mj and 10 mj is shown in Figure 9 (iii-iv).

For NCs Si composite rod, SE spectra were recorded at the same condition as NCs Si solution. The enhanced SE spectra of composite rod are quite significant as a compare to solution. The SE intensity of NCs Si in solid matrix environment at different pump energy is significantly improved as shown in Figure 10 (i).

Figure 10. Spontaneous emission spectra of NCs silicon doped sol gel (i) rod (ii) comparison solution and rod

The influence of SE intensity upon increasing pump energy from 3 mj to 10 mj is observed. When the nanocomposite rod is excited at 3mj, the intensity is quite low, but it increases dramatically at the excitation of 6mj as seen at the peaks 'b' and 'c' in Figure 10(i). Intensity is further increasing as the pump energy increases at 10 mj. No major effect at peak positions is observed upon changing the pump energy. Comparison of SE between composite sol gel and solution indicates the slightly effect on peak position. The SE peak of NCs Si in sol-gel matrix changes roughly about 10 nm from solution which is attributable to the effect of solid environment as shown in figure 10 (ii). In figure 10 (ii), the peak 'a' of NCs Si solution is slightly shifted when it is in sol-gel matrix i.e. the peaks 'b' and 'c'. Well distribution and stability of NCs Si in solid matrix environment lead to good enhancement of SE in comparison to the porous silicon solution based composite sol-gel matrix as reported in [15, 36-39]. Therefore, the obtained SE spectra of the NCs Si composite rod revealed that NCs Si are well dispersed in the matrix without aggregation. It may possible to embed high concentration of NCs Si based solution in sol-gel matrix for SE improvement that can further lead to ASE.

Conclusion

In the present work, NCs solution was prepared using NCs Si powder. The NCs Si solution was directly included in sol prepared by sol-gel technique. The NCs Si composite sol gel was further solidified to develop into rod and characterized by using different optical techniques. Well distribution and presence of NCs Si in the matrix environment were confirmed by SEM and EDX spectra, respectively. The optical properties of the composites rod are shown to be significantly stable. Photoluminescence property of NCs Si is quite enhanced in sol-gel media. Influence at peaks of NCs Si in the matrix is observed due to the solid environment. The significant spontaneous emission of the composite rod is obtained and influenced by pump energy. It may conclude that NCs siliconbased solution may directly include and stabilize in sol-gel matrix which is able to transform into a composite rod to test the stimulated emission under laser system.

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References

- [1] S. Hayashi, K. Yamamoto, Optical properties of Si-rich SiO2 films in relation with embedded Si mesoscopic particles, J Lumin*,* 70, 352-363, 1996.
- [2] L. T. Canham, Silicon quantum wire array fabrication by electrochemical and chemical dissolution of wafers, Appl. Phys. Let*,* 57, 1046, 1990.
- [3] B. S. Kim, D. I. Kim, C. W. Lee, Photoluminescence from nano silicon materials prepared by photoelectro chemical methods, J. Korean. Phys. Soc*.*, 38, 245, 2001.
- [4] U. Gosele, V. Lehmann, Light emitting porous silicon, Mater. Chem. Phys, 40, 253, 1995.
- [5] W. C. Chan, S. Nie, Quantum dot bioconjugates for ultrasensitive nonisotopic detection, Science, 281, 2016, 1998.
- [6] M. Bruchez, Jr. M. Moronne, P. Gin, S. Weiss, A. P. Alivisatos, Semiconductor nanocrystals as fluorescent biological labels, Science, 281, 2013, 1998.
- [7] T. D. Lacoste, X. Michalet, F. Pinaud, D. Chemla, A. P. Alivisatos, S. Weiss, Ultrahigh resolution multicolor colocalization of single fluorescent probes, Proc. Natl. Acad. Sci. 97, 9461, 2000.
- [8] X. Michalet, F. F. Pinaud, L. A. Bentolia, [J. M. Tsay,](http://www.sciencemag.org/search?author1=J.+M.+Tsay&sortspec=date&submit=Submit) S. [Doose,](http://www.sciencemag.org/search?author1=S.+Doose&sortspec=date&submit=Submit) [J. J. Li,](http://www.sciencemag.org/search?author1=J.+J.+Li&sortspec=date&submit=Submit) [G.](http://www.sciencemag.org/search?author1=G.+Sundaresan&sortspec=date&submit=Submit) [Sundaresan,](http://www.sciencemag.org/search?author1=G.+Sundaresan&sortspec=date&submit=Submit) [A. M. Wu,](http://www.sciencemag.org/search?author1=A.+M.+Wu&sortspec=date&submit=Submit) [S. S. Gambhir,](http://www.sciencemag.org/search?author1=S.+S.+Gambhir&sortspec=date&submit=Submit) [S. Weiss,](http://www.sciencemag.org/search?author1=S.+Weiss&sortspec=date&submit=Submit) Quantum dots for live cells, in vivo imaging, and Diagnostics, Science, 307, 538, 2005.
- [9] J. K. Jaiswal, S. M. Simon, Potentials and pitfalls of fluorescent quantum dots for biological Imaging, Trends Cell Biology*,* 14, 49, 2004.
- [10] M. N. Khan, A. S. Al Dwayyan, Influence of solvent on the physical and lasing properties of dye- doped sol gel host, J. Lumin. 128, 1767, 2008.
- [11] S. Yang, W. Cai, G. Liu, H. Zeng, P. Liu, Optical study of redox behavior of silicon induced by laser ablation in liquid, J. Phys. Chem*. C,* 113, 6480, 2009.
- [12] V. Parkhutic, Porous silicon mechanisms of growth and applications, Solid State Electron*,* 43, 1121, 1999.
- [13] D. Dimova-Malinovska, Application of stain-etched porous silicon in light emitting diodes and solar cells, J. Lumin*,* 80, 352, 1999.
- [14] A. W. Fang, R. Jones, H. Park, O. Cohen, M. Paniccia, J. E. Bowers, Integrated AlGaInAs-silicon evanescent race track laser and photodetector, *Opt. Express,* 15, 2315, 2007.
- [15] M. N. Khan, M.M. A Khan, A.S. Al Dwayyan, J.P. Labis, Comparative Study on Electronic, Emission, Spontaneous Property of Porous Silicon in Different Solvents, J. Nanomater. Article ID 682571, 2014.
- [16] L. Pavesi, Routes towards a silicon-based laser, Mater. Today*,* 8, 18, 2005.
- [17] Lalic N, J. Linnros, Light emitting diode structure based on Si nanocrystals formed by implantation into thermal oxide, J. Lumin*.* 80, 263, 1998.
- [18] R. J. Walters, G. I. Bourianoff, H. A. Atwater, Field-effect electroluminescence in silicon nanocrystals, Nat. Mater, 4, 143, 2005.
- [19] Z.X. Ma, X.B. Liao, W.C. Cheng, G.Z. Yue, Y.Q. Wang, G.L. Kong, Annealing behaviors of photoluminescence from SiOx:H, J. Appl. Phys. 83, 7934, 1998.
- [20] Y.Q. Wang, G.L. Kong, W.D. Chen, H.W. Diao, C.Y. Chen, S.B. Zhang, X.B. Liao, Getting high-efficiency photoluminescence from Si nanocrystals in $SiO₂$ matrix, Appl. Phys. Lett. 81, 4174, 2002.
- [21] T. Shimizu-Iwayama, K. Fujita, S. Nakao, K. Saitoh, T. Fujita, Visible photolumi- nescence in Si+implanted silica glass, J. Appl. Phys*.* 75, 7779, 1994.
- [22] N. Lalic, J. Linnros, Light emitting diode structure based on Si nanocrystals formed by implantation into thermal oxide, J. Lumin, 80, 263, 1999.
- [23] J.G. Zhu, C.W. White, J.D. Budai, S.P. Withrow, Y. Chen, Growth of Ge, Si, and SiGe nanocrystals in $SiO₂$ matrices, J. Appl. Phys. Vol. 78, 4386, 1995.
- [24] V. Vinciguerra, G. Franzò, F. Priolo, F. Iacona, C. Spinella, Quantum confinement and recombination dynamics in silicon nanocrystals embedded in $Si/SiO₂ super-lattices, J. Appl. Phys. 87, 8165, 2000.$
- [25] F. Gourbilleau, X. Portier, C. Ternon, P. Voivenel, R. Madelon, R. Rizk, Sirich/SiO₂ nanostructured multilayers by reactive magnetron sputtering, Appl. Phys. Lett. 78, 3058, 2001.
- [26] E. Werwa, A. A. Seraphin, L.A. Chiu, C. Zhou, K.D. Kolenbrander, Synthesis and processing of silicon nanocrystallites using a pulsed laser ablation supersonic expansion method, Appl. Phys. Lett*.* 64, 1821, 1994.
- [27] R.J. Walters, G.I. Bourianoff, H.A. Atwater, Field e ff ect electroluminescence in silicon nanocrystals, Nat. Mater. 4. 143, 2005.
- [28] G. Franz, A. Irrera, E.C. Moreira, M. Miritello, F. Iacona, D. Sanfilippo, G. Di Stefano, P.G. Fallica, F. Priolo, Electroluminescence of silicon nanocrystals in MOS structures, Appl. Phys. A: Mater. Sci. Process. A74, 1, 2002.
- [29] Y.Q. Wang, Y.G. Wang, L. Cao, Z.X. Cao, High-efficiency visible photoluminescence from amorphous silicon nanoparticles embedded in silicon nitride, Appl. Phys. Lett. 83, 3474, 2003.
- [30] L. Zhang, J.L. Coffer, T.W. Zerda, Properties of luminescent Si nanoparticles in sol gel matrices, J. Sol-Gel Sci. Technol. 11, 267–272, 1998.
- [31] V. Svrcek, I. Pelant, J. L. Rehspringer, P. Gilliot, D. Ohlmann, O. Cregut, B. Honerlage, T. Chvojka, J. Valenta, Dian photoluminescence properties of sol–gel derived SiO layers doped with porous silicon, Mater. Sci. Eng*. C*, 19, 233–236, 2002.
- [32] Y. Posada, L.S. Miguel, O. Resto, S.Z. Weisz, C.H. Kim, J. Shinar, Optical properties of nanocrystalline silicon within silica gel monoliths, J. Appl. Phys. 96, 2240, 2004.
- [33] A.Y. Karlash, Y.E. Zakharko, V.A. Skryshevsky, A.I. Tsiganova, G.V. Kuznetsov, Photoluminescence properties of silica aerogel/porous silicon nanocomposites, J. Phys. D: Appl. Phys. 43, 335, 2010.
- [34] J. Amonkosolpan, D. Wolverson, B. Goller, S. Polisski, D. Kovalev, M. Rollings, D.W.G.M. Grogan, T.A. Birks, Porous silicon nanocrystals in a silica aero gel matrix, Nanoscale Res. Lett. 7, 397, 2012.
- [35] E. Borsella, M. Falconieri, S. Botti, S. Martelli, F. Bignoli, L. Costa, S. Grandi, L. Sangaletti, B. Allieri, L. Depero, Optical and morphological characterization of Si nanocrystals/silica composites prepared by sol–gel processing, Mater. Sci. Eng, B79, 55, 2001.
- [36] A. S. Al Dwayyan, M. N. Khan, M. S. Al Salhi, Optical Characterization of Chemically Etched Nanoporous Silicon Embedded in Sol Gel Matrix, J. Nanomater, 2012, Articles ID 713203, 2012.
- [37] M. N. Khan, A. S. Al Dwayyan, M. S. Al Hossain, Morphology and Optical Properties of a Porous Silicon-Doped Sol-Gel Host, Electron. Mater. Lett*,* 9, 697-703, 2013.
- [38] M. N. Khan, A. Aldalbahi, A. alMohammedi, Investigation of Different Colloidal Porous Silicon Solutions and Their Composite Solid Matrix Rods by Optical Techniques, Journal of ELECTRONIC MATERIALS, 47, 3596, 2018.
- [39] M. N. Khan, A. Aldalbahi, A. S. Al Dwayyan, Composite rods based on nanoscale porous silicon in sol–gel silica and ormosil matrices for lightemitting applications, J Sol-Gel Sci Technol*,* 82, 551–562, 2017.
- [40] M. N. Khan, A.S. Al Dwayyan, A. Aldalbahi, Light emitting composite rods based on porous silicon in ormosils and polymer matrices for optical applications, Optics & Laser Technology*,* 9, 203–211, 2017.
- [41] M.N. Khan, A. S. Al Dwayyan, M. S. Al Salhi, M. Al Hoshan, Study on characteristics of silicon nanocrystals. within sol–gel host, J Expt. Nanosci., 7, 120, 2012.
- [42] M. N. Khan, A. S. Al Dwayyan, Influence on structural and PL property of nanocrystals silicon doped sol gel matrix, J Optoelectron Adv Mat*,* 14, 448, 2012.
- [43] M. A. Khan, S. Kumar, M. N. Khan, M. Ahamed, A. S. Al Dwayyan, Microstructure and blueshift in optical band gap of nanocrystalline Al_{x}Zn 1-xO thin, J. Lumin., 155, 275, 2014.
- [44] W. Z Yong, L. K. Xin, R. X Tang, Relative enhancement of photoluminescence intensity of passivated silicon nanocrystals in silicon dioxide matrix, Chin. Phy. B, 21, 9, 097804, 2012.