



HAL
open science

Light emission from silicon containing Sn-nanocrystals

Soren Roesgaard, Etienne Talbot, Jacques Chevallier, John Lundsgaard
Hansen, Brian Julsgaard

► **To cite this version:**

Soren Roesgaard, Etienne Talbot, Jacques Chevallier, John Lundsgaard Hansen, Brian Julsgaard. Light emission from silicon containing Sn-nanocrystals. IEEE Photonics Society Summer Topical Meeting Series, Jul 2016, Newport Beach, United States. 10.1109/PHOSST.2016.7548539 . hal-01633499

HAL Id: hal-01633499

<https://hal.science/hal-01633499v1>

Submitted on 25 Apr 2019

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Light emission from silicon containing Sn-nanocrystals

Søren Roesgaard^{*§}, Etienne Talbot[†], Jacques Chevallier^{*‡}, John Lundsgaard Hansen^{*‡}, and Brian Julsgaard^{*‡}

^{*}Interdisciplinary Nanoscience Center (iNANO), Aarhus University,

Gustav Wieds Vej 14, DK-8000 Aarhus C, Denmark.

[†]Groupe de Physique des Matériaux, Université et INSA de Rouen, UMR CNRS 6634,

av. de l'Université, 76800 Saint Etienne du Rouvray, France.

[‡]Department of Physics and Astronomy, Aarhus University,

Ny Munkegade 120, DK-8000 Aarhus C, Denmark.

[§]roesgaard@inano.au.dk

Abstract—This work investigates light emission from silicon with embedded Sn nanocrystals. The composition of the nanocrystals is determined to be pure Sn by atom probe tomography, and the light emission is strongest when the nanocrystal structure is closest to the host Si lattice diamond structure.

Index Terms—Sn, nanocrystals, photoluminescence, atom probe tomography

I. INTRODUCTION

Lately, light emission from combinations of group-IV elements has attracted special attention [1]. The available parameter space is large since the various group-IV elements can form epitaxial films in various combinations. This leads to band structures tuneable by varying the material composition and strain in the samples. Especially, GeSn alloys have received new attention since lasing was demonstrated in a waveguide structure [2]. Aside from the GeSn alloys, luminescence has also been observed from SiSn films, in form of a broad-band luminescence peak at 0.77 eV [3].

As an alternative to layers of group-IV elements or alloys, one may also investigate zero-dimensional structures such as nanocrystals. Even in the case of an indirect-band-gap material light emission would be possible due to the break-down of the momentum-conservation rule as a result of confinement [4], which is relevant in the cases of Si, Ge, and alloys containing these elements. But most importantly, the size control of nanocrystals can enable the tuning of the emission energy, making nanocrystals embedded in silicon an interesting candidate for future photonic devices. Nanostructured

silicon, in the form of porous Si or Si nanocrystals embedded in SiO₂, has been studied extensively, and the case of Ge nano-islands embedded in Si has also received considerable attention in the past. However, only limited investigations of Sn-nanocrystals have been published. Nonetheless, α -Sn, the diamond-structured allotrope of tin, is a direct-band-gap material in bulk form, and the optical transition is predicted to remain strong in the nanocrystal regime [5]. This makes Sn-nanocrystals, the topic of the present work, a very interesting candidate as a light emitter embedded in silicon.

The size of the nanocrystals can be determined by scanning transmission electron microscopy (STEM), however, one has to be careful since the final image is a convolution between the object and the electron probe size, potentially leading to mis-judgement of the particle size. Another method of size determination is atom probe tomography (APT). APT provides three dimensional mapping and the chemical nature of the atoms in the sample allowing for determination of the size and composition of individual nanocrystals.

In the present work we observe light emission around 0.85 eV from samples containing Sn-nanocrystals. APT suggests that the nanocrystals are pure Sn. The emission strength is greatly enhanced when the nanocrystals are formed in the diamond-structured phase. The nature of the light emission is discussed.

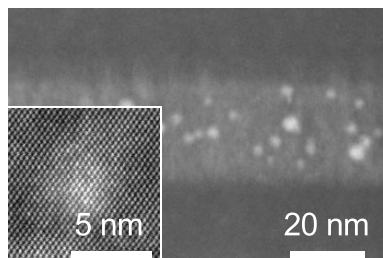


Fig. 1. STEM image of SiSnC layer annealed at 800 °C. Inset show high-resolution STEM image of a nanocrystal.

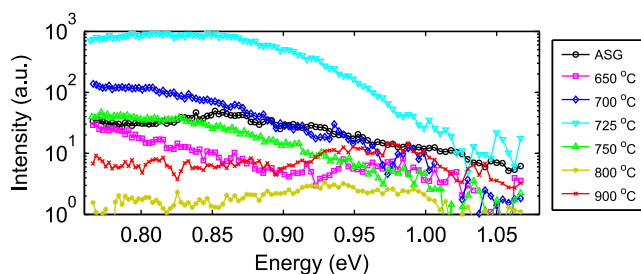


Fig. 2. Time-integrated ($0 \leq t \leq 600 \mu\text{s}$) PL spectra (please note the log scale on the vertical axis), obtained from samples annealed at different temperatures and excited by a pump fluence of $\approx 8 \cdot 10^{-6} \text{ J/cm}^2$ at a sample temperature of 16 K.

II. EXPERIMENTAL AND RESULTS

A detailed description of the sample preparation is given in [6]. In short the Sn-nanocrystal samples were prepared by molecular-beam epitaxy, where a composite layer with ratios of 1.6 % Sn and 0.04 % C where co-deposited with Si. Post-growth annealing was carried out in an N_2 atmosphere at temperatures varying from 650 °C to 900 °C for 20 min. in order to form the nanocrystals. From scanning transmission electron microscopy measurements it is known that annealing in that temperature range results in Sn-nanocrystal formation [6], as exemplified in Fig. 1.

For photoluminescence (PL) spectroscopy measurements, the samples were excited by a short laser pulse of 100 fs duration and wavelength of 400 nm (excitation energy 3.1 eV). The measurements were performed at low temperature ($T = 16$ K). Fig. 2 shows the PL spectrum for various annealing temperatures and for the as-grown (ASG) sample. A remarkably strong and rather broad luminescence feature is observed around 0.82 eV from the sample annealed at 725 °C. The center emission energy differs for the different annealing temperatures and even though part of the spectra extends below the detection limit of 0.76 eV, it is evident that the most luminescent sample is the one annealed at 725 °C. We observe this luminescence peak from low temperatures up to roughly 150 K. The crystal structure of the Sn-nanocrystals was investigated by Rutherford backscattering spectrometry using He ions. Comparing the yield of back-scattered ions under random incidence angles and under channeling conditions it is possible to extract the fraction of substitutional Sn atoms, i.e., atoms located on lattice sites corresponding to the diamond-structured host Si lattice (for more details, see [6]). This fraction is shown in Fig. 3, and we clearly observe that the enhanced light emission for the sample annealed at 725 °C correlates with the crystal structure of Sn atoms to be closest to the diamond structure, or in other words, the nanocrystals must be in the α -Sn phase.

Investigation of the chemical composition was carried out by laser-assisted three-dimensional APT, for experimental details see [7]. This technique is able to provide a spatial chemical map at the atomic scale used to determine the Sn concentration in the nanocrystals. APT results on the sample annealed at 800 °C are displayed in Fig. 4, showing a $30 \times 30 \times 20$ nm³ chemical map from the composite layer

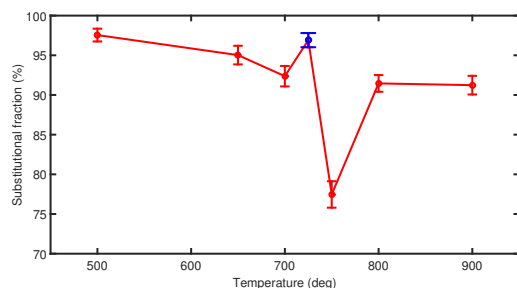


Fig. 3. Fraction of substitutional Sn atoms in relative to the Si matrix as a function of annealing temperature. The blue marks the sample annealed at 725 °C where the crystal structure of Sn atoms is closest to the Si diamond structure.

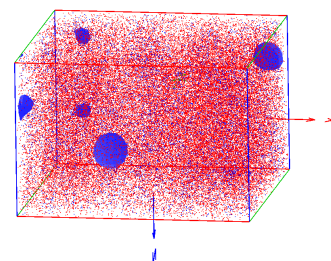


Fig. 4. $30 \times 30 \times 20$ nm³ 3D iso-concentration of the Sn (blue) and Si (red) in the composite layer.

which clearly evidences the embedded Sn-nanocrystals. The iso-concentration threshold on Sn atoms is 10 %, and the general analysis follows the methods from Refs. [8], [9]. The APT analysis suggest that spherical, pure Sn-nanocrystals are formed in the Si matrix. Moreover, not all the Sn atoms precipitate to form Sn-nanocrystals and are still diluted in the Si matrix. Intensity studies of z-contrast STEM indicate that the composition of Sn-nanocrystals is similar for the other annealing temperatures.

In conclusion light emission occurs most strongly when the Sn-nanocrystals have a structure closest to the α -Sn phase. Based on the current data, we cannot decide the precise nature of the light emission; it could occur directly from the nanocrystals, but possibly also from point defects on the nanocrystal interface or in its vicinity. However, the nanocrystals must play some role in the emission process, as the luminescence strength correlates with the nanocrystal structure.

ACKNOWLEDGMENT

This work was supported by the Villum Foundation.

REFERENCES

- [1] K. P. Homewood and M. A. Lourenço, "Optoelectronics: The rise of the GeSn laser," *Nat. Photonics*, vol. 9, no. 2, pp. 78–79, 2015.
- [2] S. Wirths, R. Geiger, N. von den Driesch, G. Mussler, T. Stoica, S. Mantl, Z. Ikonc, M. Luysberg, S. Chiussi, J. M. Hartmann, H. Sigg, J. Faist, D. Buca, and D. Grützmacher, "Lasing in direct-bandgap GeSn alloy grown on Si," *Nat. Photonics*, vol. 9, no. 2, pp. 88–92, jan 2015.
- [3] A.-S. T. Khan, P. R. Berger, F. J. Guarín, and S. S. Iyer, "Band-edge photoluminescence from pseudomorphic Si_{0.96}Sn_{0.04} alloy," *Appl. Phys. Lett.*, vol. 68, no. 22, p. 3105, 1996.
- [4] D. Kovalev, H. Heckler, M. Ben-Chorin, G. Polisski, M. Schwartzkopff, and F. Koch, "Breakdown of the k-Conservation Rule in Si Nanocrystals," *Phys. Rev. Lett.*, vol. 81, no. 13, pp. 2803–2806, sep 1998.
- [5] R. V. S. Jensen, T. Garm Pedersen, and K. Pedersen, "Optical properties and size/shape dependence of α -Sn nanocrystals by tight binding," *Phys. Status Solidi Curr. Top. Solid State Phys.*, vol. 8, no. 3, pp. 1002–1005, mar 2011.
- [6] S. Roesgaard, J. Chevallier, P. I. Gaiduk, J. L. Hansen, P. B. Jensen, A. N. Larsen, A. Svane, P. Balling, and B. Julsgaard, "Light emission from silicon with tin-containing nanocrystals," *AIP Adv.*, vol. 5, no. 7, p. 077114, jul 2015.
- [7] S. Roesgaard, E. Talbot, J. Chevallier, J. L. Hansen, and B. Julsgaard, "Scanning transmission electron microscopy and atom probe tomography of Sn nanocrystals embedded in silicon," *Unpubl. Work*.
- [8] E. Talbot, R. Lardé, F. Gourbilleau, C. Dufour, and P. Pareige, "Si nanoparticles in SiO₂: An atomic scale observation for optimization of optical devices," *Europhysics Lett.*, vol. 87, no. 2, p. 26004, jul 2009.
- [9] E. Talbot, R. Lardé, P. Pareige, L. Khomenkova, K. Hijazi, and F. Gourbilleau, "Nanoscale evidence of erbium clustering in Er-doped silicon-rich silica," *Nanoscale Res. Lett.*, vol. 8, no. 1, p. 39, 2013.