

Optical Properties and Applications of Nanoscale Silicon

Adam A. Filios, Yeong S. Ryu, and Kamal Shahrabi
Farmingdale State College
State University of New York

filiosaa@farmingdale.edu, ryuy@farmingdale.edu, shahrak@farmingdale.edu

Abstract

Advances in the design, synthesis and characterization of nano-materials are expected to provide the unprecedented ability to manipulate matter at the most fundamental level, allowing the implementation of novel nanometer scale devices and systems with unique properties and of utmost technological importance. When the physical dimensions of a device are reduced to the nanometer scale, quantum phenomena become prevalent modifying the optical and electronic properties of the material. This may enable the design and manufacturing of materials with properties tailored for the particular application.

This paper examines the unique properties of silicon-based nanostructures that are fundamentally different from those of the bulk crystalline material and make nanoscale silicon attractive for use in a variety of important technological applications such as light emitting devices for optical communications, photovoltaic solar cells, and fully integrated optoelectronic circuits. Optical and electrical properties of nanoscale silicon and several techniques towards developing silicon-based nanostructures using nanotechnology tools are explored.

Introduction

Nanotechnology has emerged as one of the most rapidly developing fields with significant implications in engineering, technology, health, the environment and the society as a whole. Further advances in this interdisciplinary field will require training of scientists and engineers able to work across traditional boundaries and utilize ideas and methods from several fields such as physics, chemistry, biology and engineering in order to address challenges both in the basic understanding of the physical phenomena as well as in the engineering aspects involving fabrication, characterization, effects of nanoparticles on humans and the environment, etc.

Although nanotechnology is considered to be a modern field in science and engineering, we should not forget that the underlying principles and the investigation of matter at the most fundamental level, that is at the atomic and molecular level, has been carried out by scientists since the earliest days of the scientific discovery. The difference is that until very recently, these studies were based either on theoretical analysis or indirect experimental observations. That is, when a scientist wanted to measure a property of

matter at the atomic level, she/he would design an experiment to observe the consequences of that property at the macroscopic level, and from those consequences he/she would obtain information indirectly about the atomic properties. Very rarely an atomic level property could be measured directly. Some macro-scale properties were measured, and from those results the atomic properties were inferred. With the tremendous progress in the engineering and technology of Electron Microscopy over the last 10 – 15 years, now, for the first time in the history of science, we are able to observe directly the atomic and molecular arrangements of matter. Modern High-Resolution Transmission Electron Microscopy (TEM) provides the capability to see directly individual atomic layers in a material, achieving resolution of a few tenths of a nanometer, which would be unthinkable only a few years ago. In addition to allowing the observation of matter directly at the atomic level, these modern nanotechnology tools provide the capability to manipulate individual atoms as well. Although still at the experimental stage, scientists have succeeded in positioning individual atoms with atomic-scale precision by using the tip of a Scanning Tunneling Microscope (STM) [1,2]. Other more established techniques such as Molecular Beam Epitaxy (MBE) and Atomic Layer Epitaxy (ALE) have been extensively used over the past 30 years to fabricate semiconductor devices atomic layer by atomic layer. For example, modern telecommunication lasers utilize very thin multilayer structures of InGaAsP/InP or GaAs/AlGaAs. Vertical Cavity Surface Emitting Lasers (VCSELs) consist of superlattices and quantum wells typically grown by MBE. Strain layer superlattices of Si/SiGe are being considered for use in high-speed electronic devices. In the traditional electronic devices such as Metal Oxide Semiconductor Field Effect Transistors (MOSFET) and Bipolar Junction Transistors (BJT), the electrons are treated as classical particles. For these large-scale devices, it is not necessary to consider the wave nature of the electrons and other quantum mechanical properties, because only the statistical behavior of a very large number of electrons plays a role. However, for nanoscale devices consisting of very thin superlattice layers or of quantum dots only a few nanometers in diameter, the wave nature of electrons plays a significant role in their electronic and optical properties. Therefore, as commercial electronic devices move into the nanoscale regime, it will become necessary to investigate and understand the quantum properties of these devices. In addition, understanding of those characteristics may lead to the ability of scientists and engineers to design and manufacture materials and devices tailored for the particular application, circumventing restrictions imposed by the natural properties of the bulk material.

The Importance of Silicon

Semiconductor devices are key components in modern electronic systems. Silicon and gallium arsenide with its related III-V compounds form the basis of the most commonly used semiconductor materials. However, silicon is by far the major player in today's electronics market, dominating the microelectronics industry with about 90% of all semiconductor devices sold worldwide being silicon based.

But why is silicon still the most important semiconductor material, especially if one considers that its own electronic properties are rather mediocre? Its dominance is mostly

due to the following aspects, which provide benefits that other electronic materials cannot easily match:

- Silicon possesses two of the most outstanding natural dielectrics, silicon dioxide (SiO_2) and silicon nitride (Si_3N_4), which are essential for device formation. In particular, SiO_2 , which is the basis of the metal – oxide – semiconductor devices (MOS) can be grown thermally on a silicon wafer, it is chemically very stable and can achieve a very high breakdown voltage. The interface defects of the thermally grown SiO_2 by reaction of oxygen with a silicon wafer are several orders of magnitude lower than those of any deposited film.
- Silicon is non-toxic, relatively inexpensive (silicon comprises about 26% of the earth's crust which makes it second in abundance only to oxygen), easy to process (a very well established industrial infrastructure in silicon processing exists around the world), and has quite good mechanical properties (strength, hardness, thermal conductivity, etc.).

For all the above reasons, silicon is the cornerstone material in electronic systems. However, one of the most important limitations of bulk silicon is in optoelectronic applications, because of its inefficiency at emitting light. This is due to its indirect fundamental energy bandgap, which essentially makes optical transitions in the bulk material at room temperature a very rare phenomenon. In a semiconductor with an indirect fundamental energy bandgap, the maximum of the valence band and the minimum of the conduction band are found at different locations in the k-space. Recombination by a single photon – which carries negligible momentum – is not allowed, because of momentum conservation. Participation of a phonon with the right momentum is necessary to satisfy momentum conservation. Phonons are quantized modes of lattice vibrations that occur in a solid. In the bulk material, this phonon assisted optical transition is very weak, allowing many other non-radiative processes to dominate resulting in a huge drop in the light emission efficiency. Bulk silicon is therefore not suitable for the implementation of optoelectronic devices. To date, the semiconductor optoelectronics industry has been dominated by the III-V compound semiconductors, because of their high efficiency in optical transitions primarily due to their direct fundamental energy band-gap.

Silicon Optoelectronics and Nanotechnology

The growth of the optical communications and related industries has generated a high demand for efficient and low-cost materials to be used for functions such as light emission, detection and modulation. In addition, silicon-based materials with improved optical properties will find important applications in enhancing the efficiency of photovoltaic solar cells, which is a market also dominated by silicon, and which is expected to experience a tremendous growth in the near future. The importance of developing a technology that would allow optical and electronic devices to be easily integrated on a silicon wafer has long been recognized. Over the past 15 years, considerable efforts are being carried out within the research community to achieving this goal. Several materials and methods have emerged as possible contenders for silicon-

based optoelectronics. These include silicon-based superlattices and quantum dots facilitating quantum confinement in silicon nanocrystals [3,4]; SiGe and SiGeC devices [5]; silicon devices doped with optically efficient rare earth impurities such as erbium [6, 7]; direct integration of III-V materials on silicon [8]; porous silicon [9, 10,]; silicon and carbon clusters embedded in oxide or nitride matrices [11]; superlattices of epitaxially grown silicon with adsorbed oxygen [12, 13].

Most of the above mentioned techniques involve devices that are based on nanoscale silicon. In a nanostructure, electrons in the conduction band and holes in the valence band are confined spatially by potential barriers. In the case of a quantum dot, carriers are confined in all three dimensions (3D quantum confinement). In a nano-wire, the carriers are confined in two dimensions and are free in only one dimension (2D quantum confinement). In a superlattice, carriers are confined in only one direction and free to move on the plane (1D quantum confinement). Such quantum confined superlattices based on gallium arsenide (GaAs) and indium phosphide (InP) have already found commercial applications in semiconductor distributed feedback lasers (DFB), semiconductor optical amplifiers, and VCSELs for optical communications [14, 15]. Fundamentally, in all cases quantum confinement pushes up the allowed energies effectively increasing the bandgap. The up-shift of the quantum confined bandgap increases as the nanoparticle size becomes smaller. It also increases as the characteristic dimensionality of the quantum confinement increases (from 1D to 2D to 3D). Therefore, quantum confinement may be used to tune the energy of the emitted light in nanoscale optical devices based on the nanoparticle size and shape.

The other important fundamental issue is the requirement for momentum conservation in the optical transition. In silicon, even for the highest degree of confinement and nanoparticle size around 3 nm in diameter, the bandgap still remains indirect. Therefore, participation of a phonon with the right momentum is required to facilitate momentum conservation in the radiative transition. The light emission in indirect bandgap silicon nanocrystals can be explained in terms of phonon assisted exciton recombination across the bandgap. An exciton is a pair of an electron and a hole bound to each other by Coulomb interaction. The exciton is in a way analogous to a hydrogen atom, but the binding energy of the exciton is much smaller than that of the hydrogen atom. In a defect free crystal, at room temperature, there are two competing processes involving phonons and excitons. One is the process of phonon assisted radiative recombination and the other the process of exciton break-up due to the interaction with phonons. In order for a phonon to participate in phonon assisted radiative recombination, it must have the right momentum to bridge the separation in momentum space between the top of the valence band and the bottom of the conduction band. However, any phonon can break-up the exciton as long as it has enough energy. In bulk silicon, the exciton binding energy is small, about 15 meV, and thermal phonons with energy $kT \approx 26$ meV have enough energy to break-up the exciton to a free electron and a free hole which move away from each other through the continuum of states in the conduction and valence bands. Therefore, exciton break-up dominates and radiative recombination becomes very unlikely. In a nanoparticle, the continuum of the valence band and conduction band states is modified into a discrete set of energy levels due to quantum confinement. Furthermore,

in a nanoparticle the exciton binding energy increases due to the confinement induced overlap of the electron and hole wavefunctions. In a silicon quantum dot of about 3 nm in diameter the exciton binding energy has been calculated to be larger than 160 meV [16], much larger than the binding energy of the excitons in the bulk as well as the energy of the thermal phonons ($kT \approx 26$ meV). Therefore, in a nanoparticle excitons cannot be broken up by thermal phonons, thus, allowing the exciton enough time to wait for the phonon with the right momentum to participate in the phonon assisted radiative recombination, producing an efficient light emission at room temperature.

Light Emission from Nanoscale Silicon

Several techniques have been explored towards developing silicon-based nanostructures utilizing quantum confinement for devices with engineered bandgap, increased functionality, and enhanced optical transitions. Porous silicon is a material produced by electrochemically etching silicon in aqueous hydrofluoric acid solutions, consisting of a network of nanometer size silicon crystallites in the form of nano-wires and nano-dots. Porous silicon exhibits bright room temperature photoluminescence in the visible region of the spectrum [9, 10]. Several models have been proposed to explain the observed luminescence, including quantum confinement in silicon nanocrystals [17], luminescence from siloxene ($\text{Si}_6\text{O}_3\text{H}_6$) and other Si-O-H compounds [18], luminescence from silicon hydride complexes (SiH) [19] and several combinations of the above models [20].

Figure 1: Photoluminescence spectra from porous silicon at room temperature (After Ref. 29).

Room temperature photoluminescence spectra of a typical porous silicon sample excited by the 457.9 nm line of an Ar ion laser are shown in Fig. 1. The vertical axis shows the relative intensity of the photoluminescence in arbitrary units (a.u.). In general, the term “arbitrary units” refers to the number of counts registered by the photodetector. The peak

energy of the photoluminescence is at 1.85 eV. The full width at half maximum (FWHM) is about 350 meV. The large width of the spectrum is attributed to luminescence from nanocrystallites with a distribution of sizes. The peak energy and the intensity of the photoluminescence are strongly correlated with preparation conditions [10]. In addition, it has been shown that the luminescence can also be tuned by various post anodization treatments such as thermal oxidation [10], or rapid thermal oxidation [21].

Several other methods for producing silicon nanoparticles have also been explored. One of the limitations of porous silicon is that the material is not very stable chemically, and that the fabrication parameters and interface chemistry cannot be fully controlled. Therefore, research efforts have been concentrated on manufacturing silicon nanoparticles utilizing standard semiconductor processing techniques such as vacuum deposition by Molecular Beam Epitaxy (MBE) or Chemical Vapor Deposition (CVD), which offer much better control in the layer dimensions and the interface quality. Devices consisting of thin silicon layers sandwiched between layers of oxide, as well as silicon quantum dots embedded in an oxide matrix have been investigated [22, 23, 24, 25]. Silicon superlattices prepared epitaxially under ultra-high vacuum deposition by MBE with silicon layers of only 1 – 2 nm thick, separated by adsorbed monolayers of oxygen have very low interface defects and show quantum confinement characteristics [12, 25]. Devices prepared from these materials showed strong photoluminescence and electroluminescence at room temperature [4, 11, 13, 23].

Figure 2: Room temperature electroluminescence (EL) and photoluminescence (PL) spectra from nanoscale silicon-based superlattices (After Ref. 11).

Figure 2, taken from Ref. 11, shows an example of room temperature photoluminescence (PL) and electroluminescence (EL) spectra from such prepared silicon superlattice. Excited by the 457.9 nm line of the Ar ion laser, the photoluminescence shows peak photon energy at 2.2 eV. The electroluminescence spectrum shows a main peak with

photon energy of 2.0 eV and extends in the blue region with a noticeable peak around 2.8 eV. Note that the blue component is absent in the photoluminescence spectra, because the laser photon energy used for excitation (457.9 nm) is not high enough to excite this transition. The vertical axis shows the relative intensity of the photoluminescence in arbitrary units (a.u.).

The measured diode current versus applied voltage of an electroluminescent sample is shown in Fig. 3, taken from Ref. 11. The device size is about 0.5 mm × 1.0 mm.

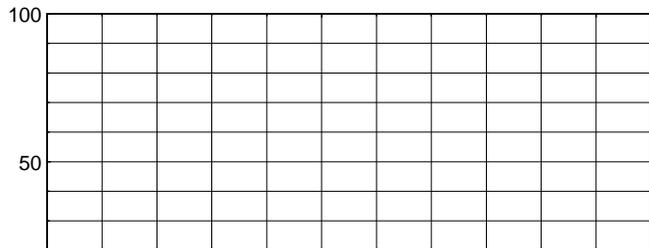


Figure 3: Diode current versus applied voltage (After Ref. 11).

Characterization of Silicon Nanoparticles by Raman Spectroscopy

From the above discussion, it becomes clear that one of the most important issues in the implementation of nanoscale devices is the ability to control the nanoparticle size and shape during the manufacturing process. One technique that can be used to indirectly measure the nanoparticle dimensions and crystalline structure in silicon is based on Raman spectroscopy.

Raman spectroscopy involves inelastic scattering of light from a material. In Raman scattering experiments, monochromatic light, typically from a laser, interacts with the lattice vibrations (i.e. the phonons) in a semiconductor, resulting in the energy of the scattered light being shifted up or down relative to the original laser line. This shift in energy provides information about the properties of the material. The three phases of silicon, (i.e. amorphous silicon, crystalline bulk silicon, and nanoscale silicon), exhibit distinct signatures in their Raman spectra. For crystalline silicon, a sharp line is seen in

the Raman spectra with a natural line-width of about 3 cm^{-1} . This sharp line for crystalline silicon is shown in Fig. 4, and it is shifted by 522 cm^{-1} relative to the laser line.

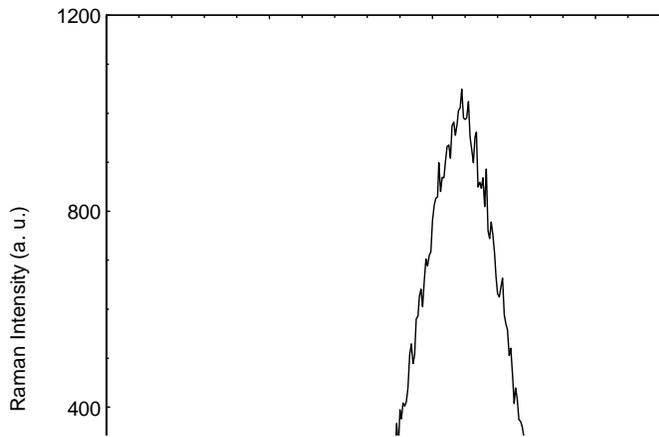


Figure 4: Raman spectra from crystalline silicon (After Ref. 29).

Amorphous silicon exhibits a very broad peak at 480 cm^{-1} relative to the laser line. Typical Raman spectra of amorphous silicon is shown in Fig. 5. Raman spectra from nanocrystalline silicon is red-shifted and broadened relative to the 522 cm^{-1} sharp Raman line for bulk crystalline silicon. This Raman shift is attributed to the quantum confinement of the electronic wavefunction in silicon nanocrystals and can provide an estimate of the characteristic dimensions of the nanocrystalline structures [10]. A correlation of the photoluminescence and Raman spectra for several types of nanocrystals has been observed [10]. As the photoluminescence peak increases in energy, the Raman peak shifts to lower energy indicating a smaller size of the nanoparticle. Typical Raman spectra from nanocrystalline silicon is shown in Fig. 6.

Figure 7 shows the transformation of the Raman spectra for a silicon sample initially prepared at the amorphous phase and then crystallized by high-temperature thermal annealing. The as prepared sample consists of a thick silicon layer deposited on a quartz substrate at room temperature by electron beam evaporation. Since the deposition takes place at room temperature, the resulting material is amorphous silicon and the Raman spectra exhibit a broad peak at 480 cm^{-1} as shown in Fig. 7 (as prepared curve). This curve has been multiplied by 5 to fit in the scale of the figure. Then the as prepared sample is annealed at various temperatures, resulting in crystallization of the silicon layer by thermal annealing.

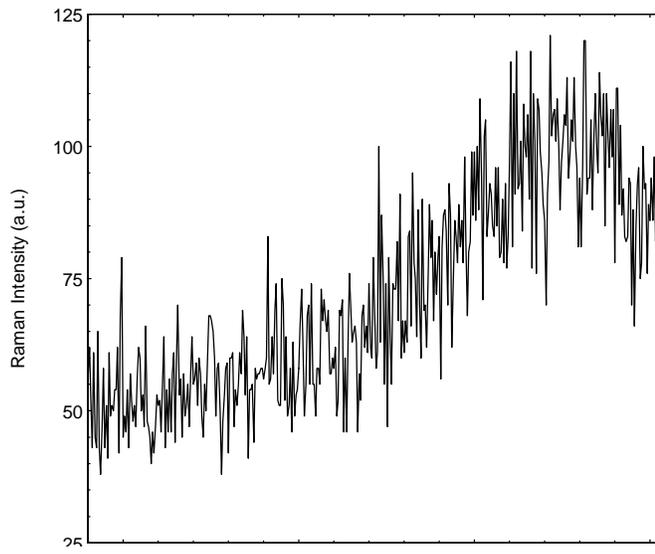


Figure 5: Amorphous silicon Raman spectra (After Ref. 29).

Figure 6: Nanocrystalline silicon Raman spectra. The red shift from the 522 cm^{-1} crystalline silicon line increases with decreasing particle size (After Ref. 29).

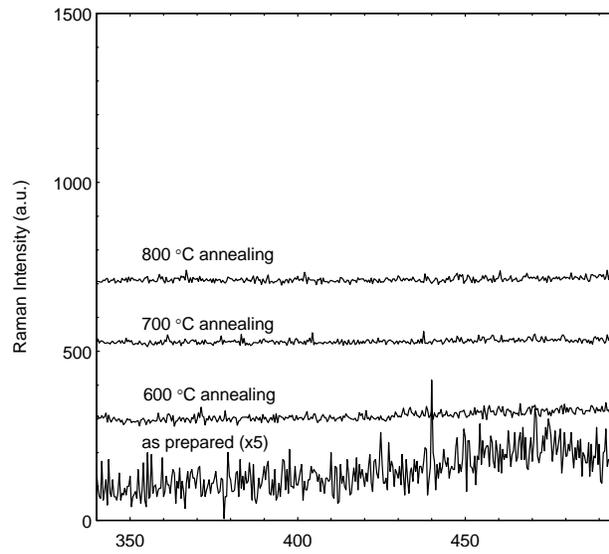


Figure 7: Raman spectra of a thick silicon layer deposited at room temperature on quartz as prepared and after annealing (After Ref. 29).

The progress of the transformation of the amorphous silicon Raman peak to the crystalline silicon peak at 522 cm^{-1} , for various annealing temperatures, namely $600\text{ }^{\circ}\text{C}$, $700\text{ }^{\circ}\text{C}$, and $800\text{ }^{\circ}\text{C}$ is shown in Fig. 7. Note that since the as deposited silicon layer is thick, there is no evidence of any nanocrystalline phase during the crystallization process. A comparison of porous silicon – which is basically nanocrystalline silicon – Raman spectra with bulk crystalline silicon Raman spectra is shown in Fig. 8.

Raman spectroscopy provides indirect information about the crystal structure. It is a rather simple and relatively inexpensive technique that can be used to obtain qualitative information. However, a more direct method is now available in the field of nanotechnology by utilizing high-resolution Transmission electron Microscopy (TEM). Although TEM is a more expensive experimental technique, it provides direct information invaluable for device design and optimization. A high resolution Transmission Electron Micrograph of crystalline silicon sample is shown in Fig. 9. Note the clear distinction of the crystalline silicon atomic layers and the interface at the top part of the figure.

Applications of Nanoscale Silicon in Photovoltaic Solar Cells

Nanocrystalline silicon films have recently attracted attention for use in photovoltaic solar cells since they show promise for providing an approach that results in lower cost and higher efficiency than conventional solar cells [26, 27, 28]. Traditionally, most of the commercially available solar cells are made out of various forms of silicon wafers.

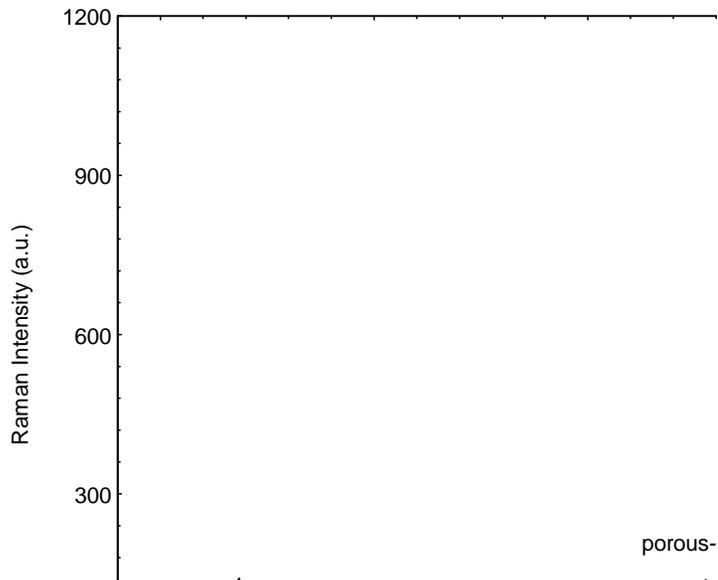


Figure 8: Raman spectra of porous silicon and bulk crystalline silicon (c-Si)
(After Ref. 29).

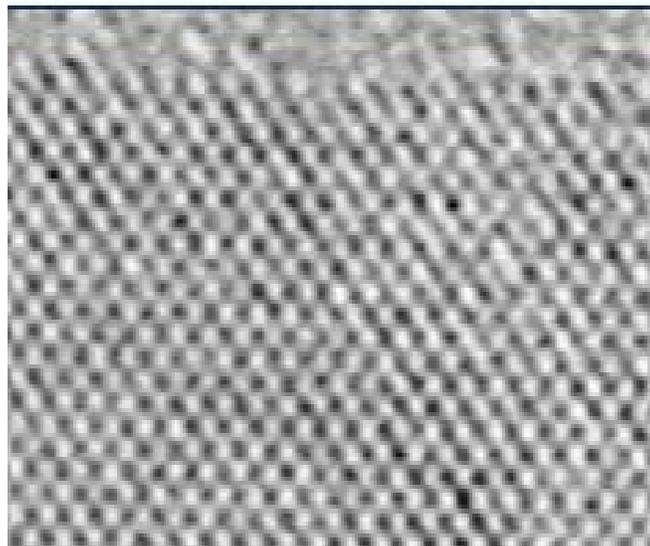


Figure 9: High Resolution Transmission Electron Micrograph of crystalline silicon.
(After Ref. 29).

Single crystalline silicon solar cells provide relatively good efficiencies, however they suffer from high cost. In addition, crystalline silicon has low absorption coefficient and narrower band gap than that desired for solar cell applications. Solar light with energy much larger than the bandgap, when absorbed by the material is converted into heat rather than electricity thus reducing the efficiency. Polycrystalline silicon or amorphous silicon solar cells are available at lower cost, however their conversion efficiency is lower than their single crystalline counterparts. In addition, amorphous silicon shows photodegradation and reduced stability under long-term illumination.

The use of nanocrystalline silicon holds the promise to realize both high performance and low cost in photovoltaic solar cells. Nanoscale silicon provides much better chemical stability than thin film polycrystalline or amorphous silicon under long-term operation. Its wider bandgap is much more suitable for solar cell applications than the narrow indirect bandgap of single crystalline silicon. Furthermore, silicon nanostructuring may provide the means for achieving a bandgap-engineered material to be used in tandem solar cells. Silicon nanoparticles of appropriate sizes or superlattices of Si/SiO₂, Si/SiGe and/or Si/C can be integrated with other materials with appropriate bandgap, such as amorphous silicon, polycrystalline silicon, or single crystalline silicon, to manufacture stacked solar cells. These types of solar cells can provide a variation of the effective bandgap across the material, allowing a significant portion of the solar spectrum to be efficiently coupled into the device and produce electricity.

Conclusion

Optoelectronic properties and characteristics of nanoscale silicon that are fundamentally different from those of bulk crystalline silicon were presented. Several techniques towards developing silicon-based nanostructures, including devices composed of silicon nano-crystallites embedded in an amorphous matrix as well as multilayer structures such as superlattices and quantum wells were briefly discussed. Potential applications of nanoscale silicon-based materials include silicon-based light emitting devices for use in optical communications, including chip-to-chip and intra-chip optical interconnects, fully integrated silicon-based optoelectronic circuits, photovoltaic solar cells, heterojunction devices, and optical sensors.

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Biographies

ADAM A. FILIOS received the B.Sc. degree in Physics from the University of Athens, Greece, in 1991, and the M.S.E and Ph.D. degrees in Electrical Engineering from the University of North Carolina at Charlotte, in 1994 and 1999 respectively. He served as an assistant professor of Engineering at Johnson C. Smith University in North Carolina for two academic years (1998 – 2000), where he taught engineering courses and conducted research in optoelectronic devices. In 2000 he joined Corning's Photonics Research and Test Center in Somerset NJ, and worked in the research and development of novel nanoscale optoelectronic devices, optical communication systems and optical fibers. From 2002 to 2006 he was a senior research engineer with Nanodynamics Inc., in New York, where he worked on wide-bandgap materials, silicon photonics, and nanoscale integrated systems. Currently he is an assistant professor at Farmingdale State College (SUNY). His research interests include nanoscale optoelectronic devices, photovoltaics and fiber optic communications. He is a member of the Institute of Electrical and Electronics Engineers (IEEE) and past member of the American Physical Society (APS), the Optical Society of America (OSA), and the Materials Research Society (MRS).

YEONG S. RYU graduated from Columbia University with a Ph.D. and Master of Philosophy in Mechanical Engineering in 1994. He served as an assistant professor of Mechanical Engineering Technology at New York City College of Technology (CUNY) for three academic years (2003 – 2006), where he taught mechanical engineering courses and conducted research in nanotechnology. In addition, he has conducted various research projects at Xerox Corporation (1994-1995), Hyundai Motor Corporation (1995-1997), and New Jersey Institute of Technology (2001-2003). Currently he is an assistant professor at Farmingdale State College (SUNY). He has been teaching and conducting research in a broad range of areas of system identification and control of nonlinear mechatronic systems and vibrations in structures requiring precision pointing to eliminate the detrimental effects of such diverse disturbance sources. He has authored or co-authored more than 70 publications. His work focuses currently on the development and implementation of modeling and control of human biology systems. He is a member of the American Society of Mechanical Engineers (ASME), and the American Society for Engineering Education (ASEE).

Dr. Kamal Shahrabi received the B.S. degree in Electrical Engineering from New Jersey Institute of Technology, Newark, NJ, in 1983, the M.E. Degree in Electrical Engineering from City College in New York, New York, NY, in 1985 and M. Phil. and Ph.D. degree in Electrical Engineering from City University Of New York, New York, NY, in 1992. Kamal Shahrabi is a Dean of School of Engineering Technologies and Professor of the Department of Electrical and Computer Engineering Technology. He has been a professor in the Technology Department at Kean University of New Jersey since 1986-2007 and has been a Chairperson of the department since September 1998-2007. Dr. Shahrabi has published several papers in various areas of Communication, Robot Tracking in Space, Wireless Communication, Impact of Globalization of Advanced Telecommunication Technologies in Developing Countries, and High Speed Optical Satellite Communication and Femtosecond Pulse Shaping. Dr. Shahrabi has several years

of experience in the areas of digital signal processing, space tracking, communication and satellite control systems while doing research at City University of New York for NASA.

In addition, he has worked as a Visiting Professor in the Electrical Engineering Department with Institut Teknologi Tun Hussein Onn, Parit Raja, Batu Pahat, Johor, Malaysia and he developed an engineering curriculum with DSP specialization for the institute (ITHO) during his stay.

His research interests are in the fields of Digital Signal Processing Algorithms, Spread Spectrum, nanotechnology, and Wireless Communication Systems.