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Photoluminescence studies on porous silicon/polymer heterostructure

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Abstract

Hybrid devices formed by filling porous silicon with MEH-PPV or poly [2-methoxy-5(2-ethylhexyloxy-*p*-phenylenevinylene)] have been investigated in this work. Analyses of the structures by scanning electron microscopy (SEM) demonstrated that the porous silicon layer was filled by the polymer with no significant change of the structures except that the polymer was infiltrated in the pores. The photoluminescence (PL) of the structures at 300 K showed that the emission intensity was very high as compared with that of the MEH-PPV films on different substrates such as crystalline silicon (c-Si) and indium tin oxide (ITO). The PL peak in the MEH-PPV/porous silicon composite structure is found to be shifted towards higher energy in comparison with porous silicon PL. A number of possibilities are discussed to explain the observations.
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Keywords: Porous silicon; MEH-PPV; Heterostructure; Photoluminescence; Exciton; Quantum confinement

1. Introduction

Organic semiconductors have attracted much attention in these days due to the solution-based processing technology by which the cost can be reduced when compared with the conventional semiconductors. Devices like polymer light-emitting diodes, photo-voltaic cells, solution-based light-emitting devices [1], flat panel display, etc. have already achieved practical success, however, the major challenge is with the substrate. In case of light-emitting diodes, generally, indium tin oxide (ITO) is used as a substrate/electrode due to its transparency as well as conductivity. Traditionally, a conducting polymeric active material is placed between an anode (generally ITO) and a cathode (may be a metal) and with suitable biasing, electrons from cathode and holes from anode are injected into the active material, which on recombination gives off the emission. Though ITO has an enormous potential and is widely used, it is not possible to integrate with the present day integrated circuits (ICs). At this point, it will be very interesting to study the silicon as an alternative substrate for polymer light-emitting devices.

There are two optical transitions associated with the fundamental absorption in semiconductors [2]: (i) direct band-to-band transition and (ii) indirect band-to-band transition. In a direct semiconductor, the conduction band minimum and valence band maximum occur at the same value of k ($k = 0$). In an indirect semiconductor, the conduction band minimum and valence band maximum occur at different points in k-space. Thus a transition from the conduction band minima to the valence band maxima requires some change in k, that is, the electron must undergo a momentum change as well as change in its energy. For example, it may go through some defect state within the band gap. In an indirect transition, the energy is generally given up as heat to the lattice rather than as an emitted photon. So, crystalline Si (c-Si), due to its indirect bandgap, is not used as an LED or laser. However, porous Si (po-Si), which is formed by an electrochemical etching of c-Si, showed luminescence. The c-Si as well as amorphous Si do not exhibit such luminescence. The po-Si exhibits luminescence at a range of wavelengths from the near infrared to the visible (blue). The wavelength depends on the thickness of the pores and the chemical state of their surfaces.

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Some works related to the filling up of the pores in po-Si by polymers were reported by different authors. Halliday et al. [3] observed an orange photoluminescence (PL) band due to recombination in the Si quantum structures both before and after coating po-Si with polyaniline. Nguyen et al. [4] studied the optical properties of po-Si/poly (*p* phenylene vinylene) devices and suggested that a very thin film of poly *p*-phenylenevinylene (PPV) had been formed in the pores of po-Si. They assumed that the energy transfer in the samples should be from po-Si to short segments in PPV emitting the light in the blue region with increase in temperature. Lopez et al. [5] had fabricated po-Si–polymer nanocomposites and described that depending on the dielectric constant of the infiltrated polymers; the PL can be tuned to emit in different regions of the visible spectrum. He concluded that the excitonic screening due to the presence of the polymers causes the emission to blue shift as much as 222 meV. They have used polymers with varying dielectric constant such as polyvinylidene fluoride (PVF), polyacrylonitrile (PAN), polymethyl methacrylate (PMMA), polyvinyl chloride (PVC), polystyrene (PS), and a fluorinated terpolymer (THV). Conwell et al. [6] has theoretically studied the interchain PL in poly(phenylene vinylene) derivatives and calculated the interchain C–C distance of 4.10–4.16 Å for poly(1-methoxy-4-(2-ethylhexyloxy-2, 5-phenylenevinylene) (MEH-PPV). Matveeva et al. [7] studied the growth of polyaniline films on po-Si layers and commented that polymerization reaction starts at the pore bottoms and then propagate towards the external surface of the po-Si layer. Recently, Levitsky et al. [8] has studied conjugated polymers entrapped in po-Si microcavity as optical sensors for low-volatility explosives such as trinitrotoluene. They reported that exposure of the po-Si microcavity containing entrapped polymer explosives vapor results in a red shift of the resonance peak. Though a large amount of works have been done on the po-Si/polymer composites, reports on the use of different substrates, such as c-Si or ITO along with the po-Si and their relative comparison are lacking in the literature. However, apart from its better solubility in solvent, its valence and conduction bands are close to the work functions of the metals used for electrodes. Such characteristics make MEH-PPV very favourable to charge injection and radiative recombination in optoelectronic devices. Furthermore, it is already well established that the morphology of MEH-PPV films can be manipulated using different organic solvents, concentrations, and spin speed, which enable control on the varying optical and electrical properties of the organic-thin-film light-emitting devices [9].

In the present study, po-Si was capped with a polymer known as MEH-PPV by spin coating. This heterostucture was studied by PL spectroscopy at 300 K and scanning electron microscopy (SEM). The PL characteristics of this MEH-PPV/po-Si heterostructure were compared with that of the MEH-PPV/c-Si and MEH-PPV/ITO to indicate that po-Si is a better candidate for emission as an alternative to ITO with additional advantage of integration with present-day IC technology.

2. Experiments

2.1. Materials

All the materials were purchased from Aldrich Chemical Company, USA, unless otherwise reported. THF, dioxane, formalin solutions, methanol, were purchased from E. Marck, India.

2.2. Preparation of MEH-PPV

MEH-PPV was prepared from 2,5-bis(chloromethyl)-methoxy-4-(2-ethylhexyloxy)benzene according to the reported procedure [10]. The product was characterized by IR, ^{1}H NMR and elemental carbon and hydrogen analyses. The weight average molecular weight of the polymer was 330,000 with polydispersity index of 2.7 as was determined by a gel-permeation chromatography with respect to monodisperse polystyrene standard.

2.3. Formation of heterostructure

P-type c-Si wafer having the (1 0 0) orientation and 15 Ω cm resistivity was taken for the fabrication of po-Si. The wafer was immersed in warm trichloroethylene (TCE). Then it was immersed in acetone for 3 min. This immersion removes the TCE residue and acts as a further cleaning solvent. The wafer, then, was dipped into methanol to remove acetone residue. Finally, it was rinsed in running deionized (DI) water to remove methanol residue. To remove ionic and metallic contamination, the wafer was then immersed in freshly prepared solution of H_2O:H_2O_2:HCl (4:1:1 parts). Then Piranha etch was used to remove organic residues from the substrate. The standard acid piranha used was a 3:1 mixture of concentrated sulfuric acid (H_2SO_4) with hydrogen peroxide (H_2O_2). The wafer was soaked for 15 min in the piranha solution, and then placed entire lot (wafer and solution) under running DI water tap.

The back of the substrate was coated with aluminium by thermal evaporation. The contact formed was ohmic as the work function of aluminium is 4.28 eV and that of silicon is 4.01 eV. The fabrication of ohmic contacts frequently includes a high temperature step so that the deposited metals can either alloy with the semiconductor or the high-temperature anneal reduces the unintentional barrier at the interface. In the case of silicon, one can simply deposit a metal such as aluminium and obtain a reasonable ohmic contact. However, subsequent annealing at 300 °C in a reducing ambient such as forming gas (20:1 N_2/H_2) will further improve the contact resistivity. The temperature is chosen below the eutectic temperature of the Si/Al eutectic composition. Annealing at higher temperature causes the formation of Si/Al alloys, which in turn causes pits in the

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