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Size dependent photoresponse characteristics of crystalline Ge quantum dots based photodetectors

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ABSTRACT

We report on the size dependent photoresponse behaviour of crystalline Ge quantum dots (QDs) dispersed within the silica matrix. Our findings demonstrate an increasing nature of EQE with increase in QDs size, which could be attributed to the combined effect of Coulomb interaction of photogenerated carriers, QD/silica interface defects and electric field driven carrier separation and tunneling through the oxide barriers. In this regard, the bias dependent nonlinear response of the photocurrent has been explained on the basis of cold field emission (CFE) model. Besides, the EQE is extended (>100%) for larger sized QDs, suggesting the trapping of slower holes in Ge QDs creating a charge neutrality issue.

1. Introduction

Band gap tunability of semiconductor QDs due to quantum size effect is attractive for enhanced performance in photonic and optoelectronic devices compared to their bulk counterparts [1,2]. This has led to strong interest to develop optoelectronic devices based on QDs, compatible with the existing mature planar Si technology [3]. In this respect, Si and GeQD sare viable candidates for photodetectors in the visible-near infrared regime [4–7] for numerous applications, since their growth process within oxide or nitride matrix is compatible with CMOS technology. GeQDs are superior due to larger Bohr radius and the requirement of a relatively lower synthesis temperature as compared to Si QDs [2]. Though there are several research articles focusing on the structural, optical, and charge storage properties of GeQDs [8–11], less attention has been paid to study their photodetection properties. Recently there are reports of photoresponse in the visible-NIR region [6,7,12] using GeQDs of amorphous nature. It is noteworthy that a QD photodetector device could be realized through crystalline or amorphous QDs and their detection properties will exhibit dissimilarity due to the structural order/disorder and the subsequent disparity in the oscillator strength, spontaneous emission rate, the carrier life time etc. Along with that, the lower temperature synthesis for amorphous quantum dots accompanies with different kinds of defects, which can further introduce some different condition and ultimately the output optoelectronic properties. Cosentino et al. reported the increasing trend of EQE of the photodetector with decreasing size of amorphous Ge QDs [12]. However, the opposite trend, i.e., increasing EQE with increase in size of crystalline Si QD was observed by Kim et al. [13]. In the present letter, we report the size tunable spectral photoresponse of crystalline GeQDs based MIS photodetectors to demonstrate the increasing nature of EQE with increasing size, which is completely opposite in case of amorphous nature as reported in ref. 12. Moreover, an enhanced EQE exhibiting a gain is observed for photodetector devices with only 30 nm active layer containing large sized Ge QDs.

2. Experimental

Radio frequency sputtering technique was employed to co-sputter Ge and SiO_2 for the deposition of Ge rich silica film (thickness ~30 nm) on top of a p-Si (001) substrate. The argon pressure, RF power, and substrate temperature were fixed at 0.02 mbar, 100 W and 200 °C, respectively. Post-deposition annealing of deposited films in N_2 ambientat different temperatures viz., 800, 900 and 1000 °C for 30 min led to the formation of GeQDs of variable size and distribution. Size and crystallinity of GeQDs were investigated using a JEOL high-resolution transmission

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electron microscope (HRTEM) with 200 keV electrons. For the fabrication of MIS devices, Al-doped ZnO (~100 nm) was deposited using a shadow mask on SiO_2 layer containing GeQDs for the transparent top electrodes. The bottom electrode was made by Al deposition on back side of the p-Si substrate followed by thermal treatment at 300 °C for 2 min to form the Ohmic contact. The current-voltage characteristics of GeQD MIS devices were studied using 4200-SCS Keithley semiconductor parameter analyzer. Spectral photoresponse of the detector were measured by a Newport EQE measurement system.

3. Results and discussion

The formation of crystalline Ge nanoclusters in the silica matrix is substantiated using HRTEM image and corresponding selected area electron diffraction (SAED) pattern. The typical HRTEM image of the sample annealed at 1000 °C is presented in Fig. 1(a). The diameter and density of QDs have been estimated as ~11 ± 3 nm and ~4.2 × 10^{11}/cm^2 respectively. The same for other samples annealed at 800 and 900 °C are shown in Table 1. The SAED pattern (inset of Fig. 1(a)) exhibits circular rings, which reveal the formation of GeQDs with different orientations inside the silica matrix. The observed diffraction rings have been indexed as (111), (220), and (311) crystallographic planes for Ge. The growth mechanism of GeQDs involves the decomposition or reduction of GeO_x into Ge [14] due to lower Gibb's free energy of SiO_2 as compared to GeO_2. The size as well as the distribution of GeQDs is found to be strongly dependent on annealing temperature, as shown in Table 1. With increasing annealing temperature, the agglomeration of QDs takes place, which results in increased size with reduced density. The crystalline nature of GeQD is further confirmed from the lattice image, as shown in the inset of Fig. 1(b). Using first Fourier transform (FFT) of the lattice fringe pattern, the lattice spacing of QD is calculated to be 3.30 Å, which agrees well with the theoretical lattice spacing (3.27 Å) of Ge (111) plane.

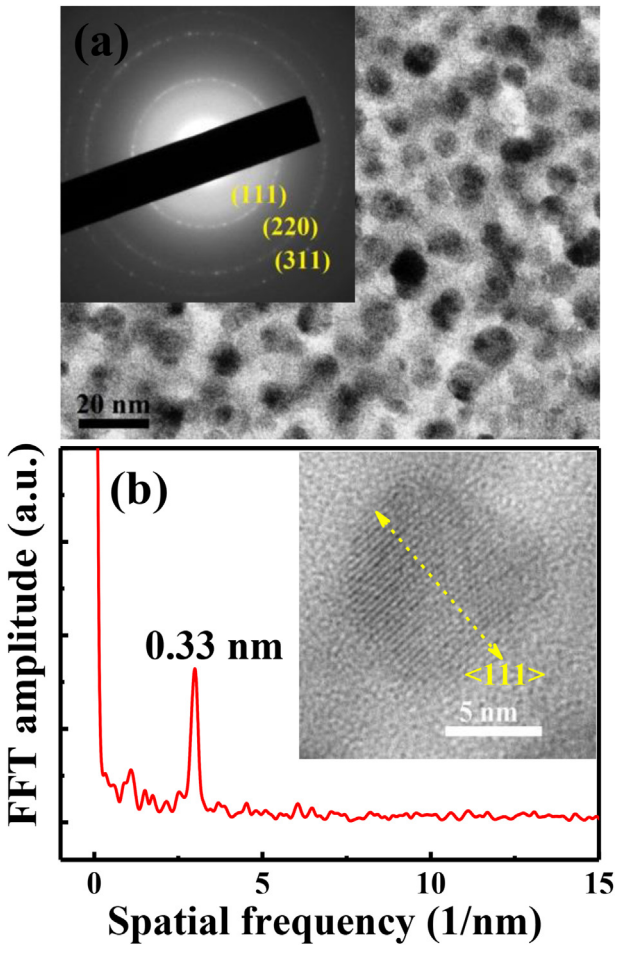

Fig. 1. (a) Typical HRTEM image of crystalline GeQDs formed at an annealing temperature of 1000 °C. Inset shows the SAED pattern from the QDs. (b) Lattice fringe pattern of typical 9 nm GeQD showing clear atomic ordering and the extracted lattice constant of 0.33 nm corresponding to (111) plane using FFT of the lattice fringe for the indicated line.

Crystalline GeQDs of varying sizes described here are expected to exhibit quantum size effect because of their diameter being well below the excitonic Bohr radius (~24 nm). The signature of the quantum effect is reflected in the size dependent relative absorbance spectra shown in Fig. 2. We have made it relative to better understand the size dependent shift. The evolution of quantum states related peaks in the absorbance spectra has been well reported for direct bandgap PbS, CdS QDs [15,16]. Also the same is true for this case. The above spectra are broad due to the size dispersion. On the other hand, the absorbance increases with increase in energy for the as-deposited film showing absence of any peak within the measured energy range. The above characteristics of GeQDs appear attractive for photodetector devices because of the broad absorption peak.

Accordingly, we have fabricated GeQDs based MIS photodetectors; a schematic structure of the device is shown in the inset of Fig. 3(a). Dark/illuminated current density-voltage (*J-V*) characteristics of G-1000 device are shown in Fig. 3(a) for different reverse bias conditions. The legend is for different excitation wavelengths corresponding to a fixed incident power density of 1 mW/cm^2, which provides an understanding of the quantum confinement induced bandgap. It may be noted that the zero bias photocurrent is very low, since a relatively high electric field is required for collecting photogenerated carriers through the oxide. However, the saturation of photocurrent takes place at a reverse bias of ~2.5 V. Applying a positive or negative bias to the substrate makes the MIS device forward or reverse biased, respectively. Photogenerated holes and electrons are separated by the reverse biased electric field and transported to the electrodes by tunneling through oxides, as schematically shown in Fig. 3(c). The spectral photoresponse at a particular reverse bias of 5 V for GeQDs annealed at different temperatures is shown in Fig. 3(b). A broad response in the spectral range 400–1100 nm could be observed, with a maximum responsivity of 0.67 A/W around 600 nm for GeQDs formed at 1000 °C (G-1000). The responsivity peak is found to be in the visible region due to transition of carriers between quantized energy levels in GeQDs. A weak peak is observed at around 1070 nm for all the devices. This response is due to the indirect band-edge supported contribution of photo-generated carriers in Si substrates.

Another important photoresponse characterization factor is the external quantum efficiency (EQE), which has been measured for different devices. The EQE measured with different reverse bias for varying QD size (2.5–11 nm) in the spectral range 400–1100 nm under continuous wave illumination are shown using a colour map in Fig. 4(a). Note that the colourbar presents the EQE values. The above figure shows that EQE values at zero applied bias are low for all devices, and increase with the increasing reverse bias. Also, the EQE values get enhanced by raising QD formation temperature, and equivalently QD diameter. For example, G-1000 device (containing GeQDs of size 11 ± 3 nm) exhibits an EQE of 130% at −5 V, whereas that for other devices (G-800 and G-900) is below 100%. An EQE above 100% signifies extra charge carrier generation in the device. The high photoconductive gain of QD detector can be explained by charge neutrality mechanism. Excitons are generated both in