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# Space charge limited current mechanism (SCLC) in the graphene oxide–Fe<sub>3</sub>O<sub>4</sub> nanocomposites/n-Si heterojunctions



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#### ABSTRACT

The graphene oxide– $Fe_3O_4$  nanoparticles ( $GO-Fe_3O_4$ ) nanocomposites/n-Si heterojunctions were fabricated and their device performance was examined through the analyses of current–voltage (I-V) and capacitance–voltage (I-V) characteristics at room temperature. It was shown that the I-V0 nanocomposites were in contact with the n-type silicon (n-Si). The seventeen different gold (I-V0 contacts performed on the I-V0 nanocomposites displayed the identical I-V0 characteristics. These results revealed that the I-V0 conditions, the ohmic and SCLC (space charge limited current) conduction behaviors were identified at low and higher voltages, respectively. Additionally, it was determined that the values of capacitance could be modified with the reverse and forward biases.

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#### 1. Introduction

Graphitic materials and their composites are hot topic research areas today [1,2]. One of graphitic materials is graphene, the oneatom-thick planar sheet of carbon atoms densely packed in a honey-comb crystal lattice [3]. It has revolutionized the scientific frontiers in nanoscience and electronics due to its excellent electrical and physicochemical properties [4–6]. Graphene is a semimetal, thus its band gap equals zero [7]. Graphene oxide (GO), the oxidized form of graphene, contains many functional groups such as hydroxyl, epoxy, and carbocyclic acid [8]. The functional groups of GO nanosheets allow a chance for their easy modification for various applications. For example, the GO sheets can be modified via doping of some functional materials such as dyes, polymers and nanoparticles, and the resultant composite materials could be used for many purposes [1,9,10]. Recently, we have reported the synthesis of GO-Fe<sub>3</sub>O<sub>4</sub> nanocomposites and their I-V and C-V characteristic of Au/GO-Fe<sub>3</sub>O<sub>4</sub> nanocomposites/p-Si/Al devices at room temperature [8].

It is known that the presence of an oxide or another interlayer between the junction materials can affect transport across the interface. This layer can also induce or inhibit interfacial chemical reactions. The current given by space charge limited current mechanism (SCLC) is observed when the contact at the junction is ohmic, where carriers can readily enter the interlayer (or insulator in MIS devices) and freely flow through the interlayer [11,12]. High density of these charge carriers causes a field gradient that limits the current density. This phenomenon mainly occurs in doped semiconducting materials. If a contact electrode is capable of injecting electrons into the conduction (or holes into the valance) band of a material, then SCLC conduction mechanism occurs. Namely, when the injection of the initial charge carrier is higher than that of recombination, the injected carriers are going to form a space-charge region and so the current flow is limited in this region. However, in the real insulators traps are distributed in accordance with the certain distribution functions and can greatly influence the current flow. In the ideal materials with no traps, the C-V characteristics displaying SCLC follow an ideal square law dependence on voltage. SCLC is the maximum current that is electrostatically allowed to flow through the device. When the current starts to flow through a junction or diode, it is initially limited by diffusion and then the current increases exponentially [13]. Finally, the current attains a space-charge limit due to the field induced by the large number of carriers present in the device. According to the SCLC mechanism, the I-V characteristics may behaves as the following: the junction device or heterojunction obeys Ohm's law for low biases, the density of carriers become larger than those of the opposite charged dopants and the device agrees with the SCLC mechanism for increased bias [14-19]. However, the mobility of carriers can still be limited by the

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interface traps. All traps are saturated at a certain voltage and then a trap-free SCLC regime in the device can be observed. In all of the current equations of different traps distributions, there is an underlying power law relationship between current and bias. The relationship is that the current density is a function of the voltage raised to a power. The linear region on  $\log I - \log V$  curve confirms SCLC mechanism. From this type of plot, the slope of the linear region or the exponent of power law relationship can be used to conclude the type of distribution of traps in interlayer materials.

In the present study, we reported the fabrication of  $GO-Fe_3O_4$  nanocomposites/n-Si heterojunctions and their I-V and C-V characteristics at room temperature. The I-V and C-V characteristics revealed that the devices had ohmic and SCLC conduction behaviors at low and higher voltages, respectively, under forward bias current-voltage (I-V) conditions.

#### 2. Experimental

#### 2.1. The preparation of n-Si

The n-type silicon wafers were cleansed by using RCA1 and RCA2 cleaning processes [8]. Organic-based contaminations were removed by the RCA1 when the RCA2 process removes metal ions on the semiconductors. The cleansing processes leaved wafers with a thin oxide film which is known that an ohmic contact is required for realizing long-lifetime operation of optical and electrical devices. If the high contact resistance occurs between the junction materials, the device performance is severely degraded through thermal stress and contact failure. After the cleansing processes, aluminum ohmic contact was executed for n-Si by vacuum evaporation at pressures of the order of 10<sup>-5</sup> Torr. The resultant n-Si/Al was annealed at 450 °C for 10 min under N<sub>2</sub> atmosphere.

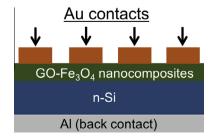
#### 2.2. The synthesis of GO-Fe<sub>3</sub>O<sub>4</sub> nanocomposites

The synthesis and characterization of GO, Fe $_3O_4$  NPs and GO-Fe $_3O_4$  nanocomposites have been reported in the literature [8]. Briefly, GO-Fe $_3O_4$  nanocomposites were synthesized by a simple liquid self-assembly method. 50 mg of GO was exfoliated in DMF (N,N-Dimethylformamide) (4.0 mL) for 2 h at room temperature in a sonicator, and then 40 mg of Fe $_3O_4$  NPs dispersed in chloroform (6.0 mL) was dropped into the GO dispersion. The final product was mixed for 3 h at room temperature in the sonicator. After the sonication process, GO-Fe $_3O_4$  nanocomposites were separated by using an external magnet.

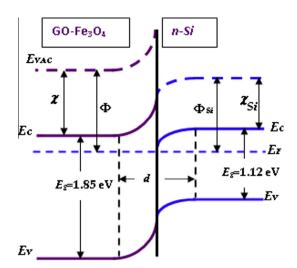
#### 2.3. The fabrication of GO-Fe<sub>3</sub>O<sub>4</sub> nanocomposites film/n-Si heterojunctions

The GO–Fe $_3O_4$  nanocomposites were coated on n-Si by using spin-coating technique. Next, seventeen Au contacts were deposited on the top of GO–Fe $_3O_4$  nanocomposites by vacuum evaporation at pressures of  $10^{-5}$  Torr. The thickness of the nanocomposites on n-Si was calculated about 165 nm from the C-V measurement. A representative schematic diagram of the fabricated Au/GO–Fe $_3O_4$  nanocomposites/n-Si heterojunctions was shown in Scheme 1.

Additionally, the energy-band diagram of  $GO-Fe_3O_4$  nanocomposites/n-Si heterojunction in equilibrium was given in Scheme 2. The band gap of  $GO-Fe_3O_4$  has been determined from the optical absorption measurements as 1.85 eV. Furthermore,  $GO-Fe_3O_4$  was found to be n-type semiconductor by the hot probe technique.



**Scheme 1.** Schematic illustration of GO-Fe<sub>3</sub>O<sub>4</sub> nanocomposites/n-Si heterojunctions.



Scheme 2. The energy-band diagram of  $GO-Fe_3O_4$  nanocomposites/n-Si hetero-iunction in equilibrium.

#### 2.4. Instrumentation

TEM (Transmission electron microscope) images were recorded by using a JEM-2100 (JEOL) instrument operating at 200 kV. Raman spectra were obtained by using a Horiba XploRA™PLUS analyses at 532.16 nm laser light. The *I–V* measurements of the heterojunctions were performed by using a KEITHLEY 487 Picoammeter/Voltage Source. The *C–V* measurements were carried out with HP 4192A, (50–13 MHz) LF Impedance Analyzer. All measurements were carried out at room temperature in dark

#### 3. Results and discussion

#### 3.1. The characterization of GO-Fe<sub>3</sub>O<sub>4</sub> nanocomposites

The details of the characterization data for GO-Fe<sub>3</sub>O<sub>4</sub> nanocomposites were given in elsewhere [8]. However, a representative TEM image of as-prepared GO-Fe<sub>3</sub>O<sub>4</sub> nanocomposites was presented in Fig. 1a. The TEM image of GO-Fe<sub>3</sub>O<sub>4</sub> nanocomposites revealed that the thin layered GO sheets were successfully decorated with Fe<sub>3</sub>O<sub>4</sub> NPs. Moreover, it is confirmed that the particle size and morphology of Fe<sub>3</sub>O<sub>4</sub> nanoparticles in the nanocomposites were maintained, which is important for material chemistry and their device applications. In addition, the formation of GO and GO-Fe<sub>3</sub>O<sub>4</sub> nanocomposites were executed by Raman spectroscopy that is a fast, non-destructive, and commonly used tool to analyses the carbon-based materials. Fig. 1b shows the Raman spectra of GO and GO-Fe<sub>3</sub>O<sub>4</sub> nanocomposites. The pristine GO shows two peaks at  $\sim\!\!1355\,\text{cm}^{-1}$  and  $\sim\!\!1600\,\text{cm}^{-1}$  which are readily attributed to D and G bands, respectively. Similar to GO, the GO-Fe<sub>3</sub>O<sub>4</sub> nanocomposites have two peaks corresponding to the D and G bands at the same Raman shifts, but with smaller intensities. Moreover, both pristine GO and GO-Fe<sub>3</sub>O<sub>4</sub> nanocomposites have the same D/G bands ratio of 0.97. Therefore, it might be concluded that the decoration of GO with Fe<sub>3</sub>O<sub>4</sub> NPs was not changed the structural properties of GO, but the presence Fe<sub>3</sub>O<sub>4</sub> NPs lowered the Raman peak intensities.

## 3.2. SCLC mechanism in GO-Fe<sub>3</sub>O<sub>4</sub> nanocomposites/n-Si heterojunctions

The current–voltage measurements are frequently used in order to determine the dominant limiting mechanism. In majority of the experimental studies, the injection mechanism is predicted from the C-V curve of the device. It is well known that the C-V characteristics of the device are affected by the presence of the traps. In an ideal material, the I-V characteristics show the

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