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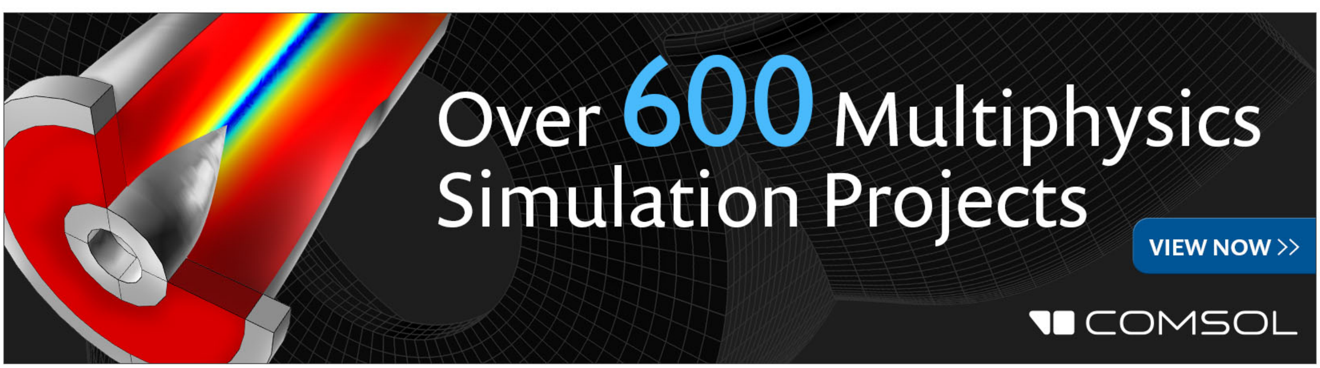

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Nitrogenated tetrahedral amorphous carbon films prepared by ion-beam-assisted filtered cathodic vacuum arc technique for solar cells application

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Fabrication and characterization of nitrogenated tetrahedral amorphous carbon (ta-C:N) semiconductor/crystalline p-type silicon (p-Si) heterojunction structures are reported. The electron-hole pairs generated from both ta-C:N and Si depletion regions were observed from photoresponse measurements. The peaks are centered at about 540 and 1020 nm, which correspond to the optical absorption edge of ta-C:N and p-Si, respectively. The reverse current increased by three orders of magnitude when the structures were exposed to AM1 light. A photovoltaic effect was observed from ta-C:N and the values of short circuit current, open circuit voltage, and field factor obtained are 5.05 mA cm^{-2}, 270 mV, and 0.2631, respectively. © *1998 American Institute of Physics.* [S0003-6951(98)02643-6]

Photoconduction behavior has been observed from diamond like carbon (DLC) films.[1–3] Two configurations have been introduced, i.e., the gap cell structure[1,2] and the heterojunction structure.[3] In the first configuration, the gaps are in the range of 10–50 μm. Light is sighted between the gaps and the photogenerated current is measured from the two contacts. A photoconductivity effect in this configuration has been reported.[1,2] However, the depletion region cannot be formed in this configuration and no photovoltaic effect has been observed. In the second configuration, with a DLC film deposited on a silicon substrate forming the heterojunction structure, the electron-hole pair excitation was observed in the Si depletion region under reverse bias as reported by Veerasamy *et al.*[3] The filtered cathodic vacuum arc (FCVA) technique was reported to be an efficient method of producing high quality, macroparticle free, and large uniformity DLC at room temperature.[3] The electronic properties were reported to be adjustable by the incorporation of nitrogen during deposition.[4] More efficient doping can be achieved by nitrogen ion bombardment during the growth of DLC films (ion-assisted FCVA).[5] In this letter, the measurements on optical absorption edge, current–voltage characteristics under dark and AM1 conditions, and photoresponse were conducted. An observation of electron-hole pairs generated from both DLC and Si depletion regions from photoresponse measurements under no bias condition is reported.

The DLC films were deposited by the FCVA technique. The substrates in the vacuum chamber were negatively biased at 80 V, which corresponds to an impinging carbon ion energy about 100 eV. The first set of ta-C films was deposited directly by a C^+ beam. The second set of samples was deposited by a C^+ beam together with a 100 eV N^+ ion beam produced by an ion beam source at a current density of 3 mA cm^{-2} and a N_2 flow rate of 2 sccm. The DLC films produced by the FCVA technique have a high sp^3 content (~88%) and are termed as tetrahedral amorphous carbon (ta-C).[6] The FCVA technique has been described elsewhere.[6] The substrates used in this study were ⟨100⟩ p-type silicon (p-Si) with a resistivity of 1–10 Ω cm. Before deposition, all substrates were cleaned with Ar ions to remove the oxide layers. Gold contacts with an area about 20 mm^2 and a thickness about 20 nm were deposited on the top of ta-C layers. The gold transmittance varied from 20% to 50% as the wavelength varied from 350 to 1100 nm.

As-deposited ta-C films exhibit a band gap of 2.7 eV[7] and a Fermi level measured by activation energy is about 0.35 eV below the midgap.[5] In this case, the films exhibit a defect-controlled p-type semiconductor characteristic. When nitrogen is incorporated in the film, the Fermi level moves up with the doping level.[5] At a nitrogen flow rate of 2 sccm, the Fermi level of the films is above the midgap, thus the films become doped n type.[5] The Fermi level is 0.8 eV below the conduction band and the band gap reduces to about 2.5 eV due to the C–N alloying effects.

The absorption coefficient (α) as a function of photon energy for the ta-C films was determined by spectroscopic phase modulated ellipsometry (UNIVEL, ISA Jobin Yvon).[7] The realization of the efficient photovoltaic energy conversion is that the film must have a sufficiently large optical absorption coefficient to absorb a significant fraction of the solar energy. As shown in Fig. 1, the optical absorption coefficient of nitrogenated ta-C (ta-C:N) is larger than that of the as-deposited ta-C. The absorption spectra for the ta-C:N are similar to those obtained from hydrogenated amorphous silicon. Each absorption spectrum shows two main absorption regions: The first is located at the interband region, which can be used to define the optical energy band gap. The second is the shoulder region, which located at the intraband region. In our previous spectral ellipsometry study (using the Forouhi and Bloomer, F. B. formulations), we can only simulate the first (interband) region.[7] However, with the Tauc and F. B. formulations, both interband and intraband regions can be clearly observed and the optical modeling for ellipsometry measurement is described elsewhere.[8] The optical band gap can be derived from the optical absorption

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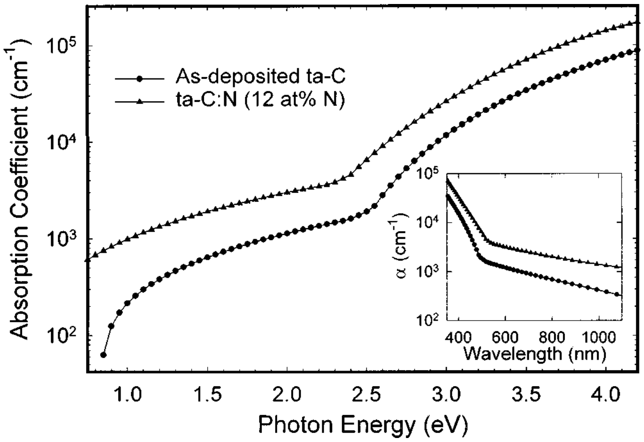

FIG. 1. Absorption spectra as a function of photon energy of the as-deposited *ta*-C film and the *ta*-C:N film with 12 at % N measured by spectral ellipsometry. (Inset shows the absorption spectra as a function of wavelength for *ta*-C and *ta*-C:N films.)

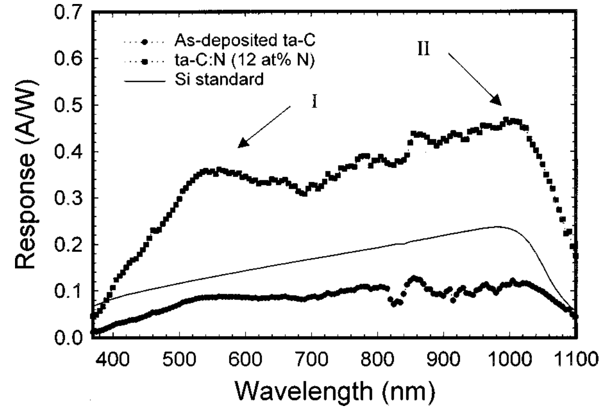

FIG. 3. Photoresponse measurements for the as-deposited *ta*-C, *ta*-C:N of 12 at % N and the Si standard.

edge.[7] The optical band gap for the as-deposited *ta*-C and *ta*-C:N are 2.7 and 2.5 eV, respectively.

The current–voltage ($I-V$) characteristics were measured with a precision semiconductor parameter analyzer (HP4156A, HP). The $I-V$ curves of the *ta*-C and *ta*-C:N under the dark and AM1 light conditions are shown in Fig. 2. The reverse bias current increases by about three orders of magnitude when the *ta*-C and the *ta*-C:N heterojunction structures are exposed to the light, i.e., the reverse bias current of *ta*-C:N heterojunction increases from 6.10×10^{-5} to 5.45×10^{-2} A cm^{-2} with a reverse bias of 2.0 V. The forward bias current was found to increase by one to two orders of magnitude. Thus, these heterojunctions are very sensitive to the light under the reverse bias conditions and are potentially useful for photodetector applications. The photovoltaic behavior was observed in the *ta*-C and *ta*-C:N heterojunction structures. Several parameters for the photovoltaic behavior, i.e., the open circuit voltage (V_{OC}), short circuit current density (J_{SC}), and field factor (FF) can be extracted from the $I-V$ measurement. The V_{OC} and J_{SC} of the *ta*-C heterojunction structures are about 0.22 V and 1.37 $\times10^{-4}$ A cm^{-2}, and the V_{OC} and J_{SC} of the *ta*-C:N heterojunction structures are about 0.27 V and 5.05 $\times10^{-3}$ A cm^{-2}, respectively. The J_{SC} of the *ta*-C:N heterojunction is about an order higher than the *ta*-C heterojunction. The FF is 0.2472 and 0.2631 for *ta*-C and *ta*-C:N heterojunction structures, respectively.

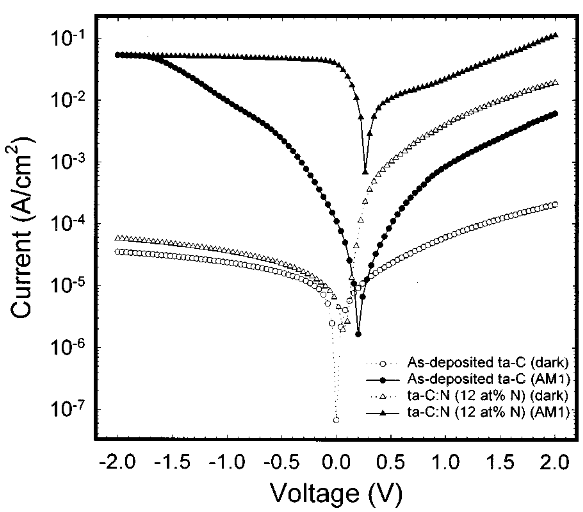

FIG. 2. $I-V$ curves for the as-deposited *ta*-C and the *ta*-C:N of 12 at % N in dark and under AM1 condition.

The photoresponse spectra of the *ta*-C and *ta*-C:N heterojunction diodes were examined by using 300 W mercury xenon arc lamp manufactured by Oriel Instruments. The wavelength of the arc lamp ranged from 350 to 1100 nm. The incident radiation was modulated in 25 Hz to reduce the signal-to-noise ratio. The spectra resolution of the system is determined by a monochromator (model 77200, 1/4 m grating, Oriel Instruments) and was fixed at 0.1 nm. The responsivity of the *ta*-C and *ta*-C:N was obtained by a comparison with a standard silicon detector head of known responsivity (Oriel's model 70331). The *ta*-C and *ta*-C:N samples were measured under no bias condition. The measurements show that the photoresponse for the *ta*-C:N materials is larger than the *ta*-C films and the standard Si detector. The noteworthy feature of the photoresponse spectra as a function of wavelength is that the both *ta*-C and *ta*-C:N spectra show two peaks centered at about 520–540 nm (band I) and 1020 nm (band II). Band I is found to coincide with the *ta*-C and *ta*-C:N optical absorption edges, as shown in the inset of Fig. 1. Therefore, it can be inferred that the photogeneration of carriers occurs in the depletion region on the *ta*-C or *ta*-C:N side of the heterojunction. The generation consists of exciting electrons and holes across the forbidden gap from a high-density valence band tail states to high-density conduction band tail states. The photoconduction takes place predominantly in this mobility edge but not the delocalized electrons or by hopping motion in the shoulder region. This agrees with our results on thermal activation measurement, which the band tail states transport mechanism is observed at room temperature.[5] Band II corresponds to the optical absorption edge of silicon. This band has a very similar trend in responsivity to the standard silicon head, as shown in Fig. 3.

In summary, the *ta*-C and *ta*-C:N heterojunction solar cells were successful fabricated. The optical absorption coefficient of these structures is sufficiently large for them to absorb a significant fraction of the solar energy. The electron-hole pair generation in the *ta*-C and the *ta*-C:N depletion regions were illustrated in the photoresponse measurements. The photovoltaic effect was observed from the *ta*-C and *ta*-C:N heterojunction structures. The analysis on the optical absorption spectra, $I-V$ characteristics under dark and AM1 conditions, photovoltaic behavior, and photoresponse spectra shows that the *ta*-C:N heterojunction structures are more promising than the *ta*-C heterojunction structures for solar cell applications.

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