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その他のタイトル	III族窒化物半導体分極制御構造による太陽光エネルギー変換に関する研究
学位授与年月日	2017-03-23
URL	http://doi.org/10.15083/00075725

論文の内容の要旨

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(III 族窒化物半導体分極制御構造による太陽光エネルギー変換に関する研究)

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Solar energy is a promising source of a renewable energy because of its huge potential. The most widely used device that harvest the solar energy is a solar cell where the solar power is converted into electricity. Water splitting photoelectrode and photocatalyst, based on the photoelectrochemical (PEC) reaction, are the other types of the promising solar energy conversion devices, in which sunlight is converted into hydrogen. Since hydrogen is much easily stored comparing to the electricity, the solar to hydrogen conversion enables us to obtain a stable power from the fluctuating solar energy. The PEC reaction, however, often suffers from the short lifetime and low energy conversion efficiency due to the complex material requirements.

III-Nitride semiconductors are potential material candidates for solar cell application because of their wide range of bandgap energy covering UV to IR (3.4 eV to 0.7 eV) and high absorption coefficient in the range of 10^5 cm^{-1}. Furthermore, III-Nitrides possess chemical stability and band edge potential that straddles the redox potentials of water oxidation and reduction reactions. Therefore III-Nitride semiconductors are also suitable for the PEC applications.

One of the big challenges lies in the growth of InGaN layers. Due to a thermal instability and a large lattice mismatch of In containing layer, the growth of high quality InGaN layer with high indium composition is still quite challenging. There is another challenge in the device structure of both solar cells and photoelectrodes. When devices are grown on +c plane which is the most common plane for III-Nitride semiconductors, the tremendously huge polarization charges often generate an electric field in the opposite direction to the carrier separation in InGaN layers. Furthermore, photoanode of III-Nitride often suffers from severe photocorrosion while photocathode often suffers from the difficulties in Mg doping and low conductivity due to low carrier mobility.

In this study, a novel device structure for +c plane, which is designed based on polarization engineering, is proposed to overcome some of the challenges in the solar energy conversion devices using III-Nitride. The proposed structure, InGaN/AlN/GaN, contains a tunnel junction induced by polarizations of a thin AlN. Owing to the unique band diagram and the tunnel junction, the direction of the carrier separation in this device is opposite to that of conventional pin structures. Therefore, the electric field in InGaN induced by the polarization charges does not prevent the carrier separation in the proposed structure. This concept has been proved by numerical calculations of the solar cell properties and nice I-V characteristics are obtained over the entire indium composition with the proposed structure.

Due to the photocurrent flow in the opposite direction, the polarization engineered structure works as a photocathode even though the structure contains no p-type layers. Therefore, the polarization engineered structure is expected to overcome the low conductivity issue of photocathode with p-type III-Nitride semiconductors. In fact, higher current density and stable operation of GaN/AlN/GaN photocathode have been experimentally demonstrated.

Despite the potential advantages of the polarization engineered structure, there exist several challenges in the growth of the GaN/AlN/GaN structure by metal organic vapor phase epitaxy (MOVPE). AlN is typically grown at higher temperature than the GaN because of the poor surface migration of Al precursors. Nevertheless, AlN grown at around 1100^{o}C have found to contain gallium (Ga) even in the absence of an intentional Ga precursor supply. This unintentional Ga incorporation is caused by the solid phase interdiffusion or due to the Ga containing deposition on the reactor wall. Thus, low temperature growth of AlN layer is quite effective to suppress the Ga incorporation and therefore provides more abrupt GaN/Al(Ga)N interfaces which are of importance for obtaining a low-resistance polarization-induced tunnel junction. The higher Al content is also desirable since the larger polarization charges induced by AlGaN makes the tunneling resistance lower. As a result, the best PEC properties have been obtained with an AlN layer grown at 800^{o}C.

The difference in the growth conditions of AlN and GaN requires growth interruptions before and after the growth of AlN layer for changing the reactor conditions. The growth interruptions, however, have been found to have a significant impact on the grown GaN or AlN layer. If the reactor conditions are changed in a conventional manner during the first growth interruption, several dot structures are formed on the surface of GaN template. The formation of dot is largely suppressed in the improved transition sequence in which higher NH_3 partial pressure is kept. The layers grown after the optimized transition of growth conditions shows improved properties compared with those grown on the GaN with dot structures.

Lattice relaxation of the AlN grown at low temperature of 800^{o}C has found to take place during the second growth interruption. Reduced piezoelectric polarization and higher threading dislocation densities due to the lattice relaxation result in the low PEC properties of the GaN/AlN/GaN photocathode. The lattice relaxation of AlN has been found to be prevented by introducing a GaN capping layer (LT-GaN) on the AlN layer before the transition of the reactor conditions. No significant degradation in crystallinity of the top GaN layer is observed when the LT-GaN layer is introduced.

Based on the growth optimizations, visible light operation of the InGaN/AlN/GaN photocathode has been successfully demonstrated. The photoresponse of the structure with 21% indium under the illumination of light with the wavelength of up to 440 nm has been obtained. The onset potential of more positive than the redox potential of water oxidation has been achieved with indium composition of up to 12.8%. The overall water splitting using the polarization engineered photocathode is, therefore, expected by further optimization of the growth conditions in the future.