



Simple method for significant improvement of minority-carrier lifetime of evaporated BaSi₂ thin film by sputtered-AlO_x passivation



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ABSTRACT

A novel surface passivation of AlO_x on BaSi₂ thin films fabricated by vacuum evaporation for solar cell application has been developed. The minority-carrier lifetime of evaporated BaSi₂ film is shorter than that of epitaxial film. This lifetime has been improved by forming AlO_x passivation layer on the BaSi₂ surface by RF sputtering of Al followed by oxidation in air. A drastic improvement of the lifetime upon the passivation of AlO_x was observed up to 15 μs with 3-nm of AlO_x layer after rapid thermal annealing at 475 °C for 15 min. The dependence of lifetime on passivation layer thickness disappeared for thickness higher than 9 nm, especially after annealing, showing the chemical passivation effect of AlO_x. The result confirmed that AlO_x is a promising passivation-layer for improving properties of BaSi₂ film.

1. Introduction

BaSi₂ is a promising material for photovoltaic thin film. It has a large absorption coefficient of $3 \times 10^4 \text{ cm}^{-1}$ at 1.5 eV, and an optical band-gap of 1.3 eV near of the ideal [1]. BaSi₂ shows n-type conductivity with electron concentration about 10^{16} cm^{-3} [2,3]. The minority-carrier lifetime, τ_m , and the diffusion length, L , are very important parameters for solar cell applications. These two parameters largely affect the short-circuit current and open-circuit voltage of a photovoltaic cell [4]. A minority-carrier diffusion length (10 μm) of as-grown film and a long minority-carrier lifetime ($> 10 \mu\text{s}$) of annealed film were observed for the undoped n-BaSi₂ prepared by molecular beam epitaxy [5–7]. However, τ_m of BaSi₂ film grown by vacuum evaporation was shorter compared with the epitaxial film. Hara et al. reported that τ_m of vacuum evaporated BaSi₂ film is about 0.02–0.6 μs [8,9], which is much shorter compared to 10 μs of the epitaxial film. It might reflect the lower quality of evaporated film than epitaxial film. By a previous DLTS study [10], only majority-carrier traps, which are located at 0.1 and 0.2 eV below the bottom of the conduction band were found in epitaxial films. However, due to the low quality of the evaporated films, the presence of minority-carrier traps is expected. Thus, we believe that the evaporated BaSi₂ film requires further treatment either during the preparation or after, such as annealing and

surface passivation to improve the minority-carrier lifetime.

Takabe et al. [7] have grown BaSi₂ epitaxial film on Si(111) substrate and characterized their minority-carrier lifetime. They concluded that the Shockley-Read-Hall lifetime, τ_{SRH} , does not depend on the average grain size, but on surface condition. The film with a mirror surface showed 0.4 μs, whereas, the cloudy or capped film by 3-nm Ba or amorphous-Si, a-Si, showed a large τ_{SRH} of 8 μs. They also have applied the a-Si layer with different thickness and air exposure time to p-type BaSi₂/n-type Si solar cells, and found that the power conversion efficiency is improved upto 9.9% [11–13]. This is considered partly due to the surface passivation effect of a-Si layer.

AlO_x is an effective material used for the passivation of silicon wafer in order to improve the minority-carrier lifetime [14–16]. The surface recombination rate of p-type Si wafer can be decreased to 9 cm/s by AlO_x passivation [14]. This passivation is based on the chemical passivation and the field effect caused by a high density of negative fixed charge at the interface between SiO₂ and AlO_x. This leads us to attempt the passivation of BaSi₂ surface by AlO_x layer.

In this paper, the effect of AlO_x passivation on τ_m of BaSi₂ thin film prepared by vacuum evaporation was studied. Al deposition layer was carried out by RF-sputtering followed by air exposure. The effect of passivation thickness and annealing on τ_m was studied. The compositional, morphology, and structural characterizations are discussed.

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2. Experimental method

2.1. Preparation of BaSi₂ thin film

BaSi₂ source (99% in purity, Kojundo Chemical Lab) was placed on a resistive tungsten boat, which was used as an evaporation element. BaSi₂ film was prepared on n-Si(111) ($\rho > 1000 \Omega \text{ cm}$). The substrate was properly cleaned by deionized water, followed by acetone, and then 5% HF acid for 10 s. After that, it was dried by nitrogen gas flow. The substrate was quickly transferred to the evaporation chamber, which is then evacuated up to 7.5×10^{-6} mbar. Then, the substrate was heated up to 550 °C and maintained at this temperature till the end of the evaporation, and then cooled down to room temperature. For BaSi₂ evaporation, the tungsten boat was heated to low temperature for 30 s before opening the shutter in order to remove any surface contamination, and then was quickly heated up to high temperature beyond the melting point of BaSi₂ and was kept for 30 s. The source weight of BaSi₂ was 0.2 g, and the remaining weight after the evaporation was about 0.05 g. The thickness of BaSi₂ film is ~650 nm, as measured by SEM.

2.2. Preparation of AlO_x passivation layer

For the passivation layer, Al film with 3 nm thick was deposited on the surface of BaSi₂ film by RF-sputtering at room temperature. Then the Al film was oxidized in air. This process was repeated for more than one cycle in order to get various thicknesses of AlO_x film. It is observed that if the deposited Al was thicker than 3 nm, the formation of AlO_x is not possible and Al remains. Fig. 1 illustrates the process of BaSi₂ and AlO_x preparation. After AlO_x preparation, the films were annealed to reduce the defects by using the rapid thermal annealing system. The films were annealed at 475 °C for 15 min in Ar atmosphere of an evacuation to 10^{-6} mbar. The annealing temperature was prescribed lower than the preparation temperature (550 °C) of BaSi₂ film in order to study only the effect of AlO_x passivation.

2.3. Characterization techniques

The film structure was characterized by X-ray diffraction analysis (XRD: Bruker Discover D8) and field emission scanning electron microscopy (FESEM: JEOL JSM-7001FA) combined with energy-dispersive X-ray spectroscopy (EDS). The Cu-K α radiation of wavelength of 1.546 Å was used for XRD. The BaSi₂ films were also characterized by Raman spectroscopy (Nanofinder, Tokyo instruments, Inc.) with an Ar⁺

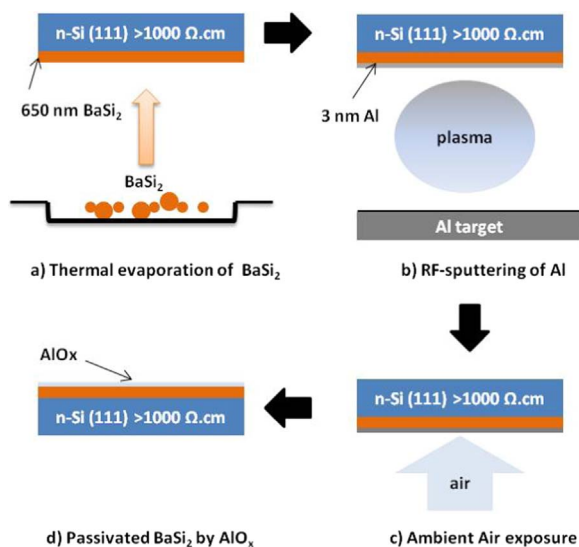


Fig. 1. Illustration scheme of BaSi₂ and AlO_x preparation.

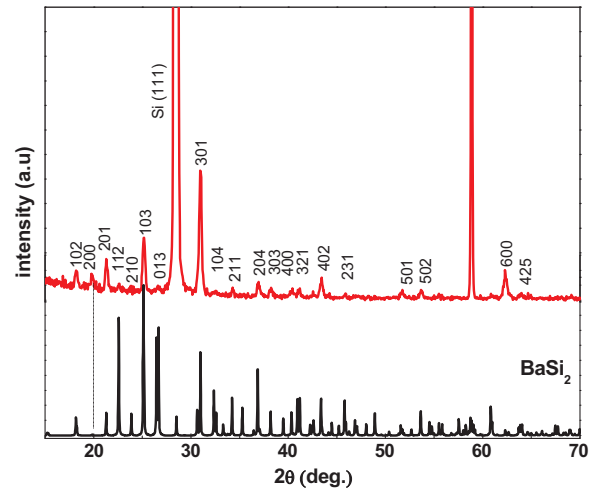


Fig. 2. XRD of the prepared BaSi₂ film at 550 °C.

ion laser ($\lambda = 488 \text{ nm}$). Excess-carrier decay kinetics was measured by microwave-detected photoconductivity decay (μ -PCD) method (KOBELCO LTA-1512EP). Excess carriers were generated by a 5 ns laser pulse with a wavelength of 349 nm. The spot size of the laser is 2 mm in diameter. Excitation-laser intensity was varied in the range of 1.1×10^2 – $1.3 \times 10^5 \text{ W/cm}^2$, corresponding to the photon density in the range of 9.8×10^{11} – $1.1 \times 10^{15} \text{ cm}^{-2}$. Photoconductivity decay was monitored by the differential detection of reflected microwave intensity with a frequency of 26 GHz.

3. Results and discussions

XRD chart of the as-prepared BaSi₂ film is shown in Fig. 2. The XRD peaks of the film are in good agreement with the theoretical pattern of the orthorhombic phase of BaSi₂ [17], and no other diffraction lines are observed than Si (111) peaks. The peak intensities of (301), (103), and (201) are markedly strong, and the preferred orientation of the film is (301).

For the passivation surface, afterward the synthesis of BaSi₂ by vacuum evaporation, it was followed by deposition of 3 nm of Al by RF-sputtering followed by air oxidation at room temperature. For deep understanding of AlO_x formation, the thickness of AlO_x layer was changed in a range 3–27 nm. Fig. 3 shows the EDX spectra of the unpassivated and passivated films. All films show Si K, Ba L, Ba M, and C K α peaks. The only passivated films showed Al K α and O K α peaks, which increased in intensity with Al thickness. The overlapped peaks in range of 0.4–0.6 keV at which the O K α and Ba M ζ existed were separated. Increasing tendency of oxygen amount is clearly observed, as seen in Fig. 4. In addition, a slight increase in oxygen amount after annealing is observed. This increase is probably attributed to the diffusion of oxygen into BaSi₂ or the oxidation of remaining Al. For further investigation of film structure, Raman spectra were measured for the films after the annealing, as shown in Fig. 5. Raman scattering is sensitive to the degree of crystallinity and surface of the sample. Five peaks are observed in the range of 200–600 cm^{-1} . The peaks are ascribed to the vibration of [Si₄]⁴⁻ anion [18,19]. These results indicate that the films contain orthorhombic BaSi₂. BaSi₂ peaks are clearly seen and not affected by the passivation or the annealing temperature.

Fig. 6 shows the effect of AlO_x passivation on the carrier-recombination kinetics at excitation level of photon density of $1.6 \times 10^{14} \text{ cm}^{-2}$. In the un-passivated film, the microwave intensity decreases rapidly due to the rapid recombination of the excess carriers. The time of the first exponential (1/e) decay ($\tau_{1/e}$) is 0.2 μs ; however, this time is much longer in case of the passivated films. In other words, with increasing AlO_x thickness, the excess carrier recombination is

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