Impact of Ba/Si Flux Ratio During Molecular Beam Epitaxy Growth on the Characteristics of BaSi$_2$ Epitaxial Films on Si(111)

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1. Introduction
Semiconducting BaSi$_2$ is considered a potential material for thin-film solar cell because of its large absorption coefficient of $3 \times 10^4$ cm$^{-1}$ at 1.5 eV, and suitable band gap of 1.3 eV. BaSi$_2$ also has a long minority-carrier lifetime of 10 μs. Therefore, we expect a conversion efficiency to be larger than 25% only in a 2-μm-thick BaSi$_2$ pn junction diode [1]. BaSi$_2$ can be grown on a Si(111) surface by molecular beam epitaxy (MBE), co-depositing Ba and Si on a heated Si substrate. In our previous research, we utilized the large Ba/Si flux ratio ($R_{Ba}/R_{Si}$) during the MBE growth because the vapor pressure of Ba is much larger than that of Si [2]. We expect that this $R_{Ba}/R_{Si}$ has significant influences on the electrical and optical properties. According to M. Kumar et al., first-principle calculations revealed that the formation energies of the point defects in BaSi$_2$ differ according to the growth conditions [3]. Hence, we anticipate that the amount of point defects and their species in BaSi$_2$ change with varying $R_{Ba}/R_{Si}$. In this work, we grew undoped BaSi$_2$ epitaxial films with different $R_{Ba}/R_{Si}$ and investigated the effect of $R_{Ba}/R_{Si}$ on the crystalline qualities and optical properties.

2. Experiment
We fabricated 500-nm-thick undoped BaSi$_2$ on Si(111) substrates at 580 °C by MBE. We used low resistivity n-Si(111) ($\rho = 0.01$ Ω·cm) for the photoresponsivity measurement and high-resistivity p-Si(111) ($\rho = 1000 - 10000$ Ω·cm) for the Hall measurement. The deposition rate of Si during the MBE growth $R_{Si}$ was fixed at 0.9 nm/min and that of Ba $R_{Ba}$ was varied from 0.9 to 3.6 nm/min, that is, $R_{Ba}/R_{Si}$ from 1.0 to 4.0. After growing the samples, we formed a 3-nm-thick amorphous Si passivation layer on the BaSi$_2$ surface at 180 °C [4]. Finally, we sputtered 1-mm-diameter and 80-nm-thick indium-tin-oxide electrodes on the front and 150-nm-thick Al electrodes on the back surfaces at room temperature. The real atomic ratio $N_{Ba}/N_{Si}$ of the films was measured by Rutherford back scattering (RBS) measurements. We measured the internal quantum efficiency (IQE) spectra at RT, and convert IQE to the light current density ($J_L$) by using $J_L = q \times \Phi_{AM,1.5} \times IQE \times dE$. The carrier concentration was evaluated by Hall measurement using the van der Pauw method and/or capacitance versus voltage (C–V) characteristics.

3. Results and Discussion
Figure 1 shows $N_{Ba}/N_{Si}$ depth profiles of the grown BaSi$_2$ films with $R_{Ba}/R_{Si}$ of 2.2 and 4.0. The $N_{Ba}/N_{Si}$ value of the sample with $R_{Ba}/R_{Si} = 4.0$ was entirely larger than that with $R_{Ba}/R_{Si} = 2.2$. In addition, the $N_{Ba}/N_{Si}$ decreased when it approaches the BaSi$_2$/Si interface. We thought this is because the Si substrates supplied the Si atoms to the BaSi$_2$ layer during the MBE growth. Figure 2 shows the dependence of $J_L$ and the carrier concentration on $R_{Ba}/R_{Si}$. The $J_L$ reached a maximum and the carrier concentration reached a minimum when $R_{Ba}/R_{Si} = 2.2$. Moreover, BaSi$_2$ films with $R_{Ba}/R_{Si} = 2.2$ only shows p-type conductivity with a hole concentration of $1 \times 10^{15}$ cm$^{-3}$. According to ref. [3], $V_{Si}$, $B_{Si}$, and $S_{Si}$ are easily generated in BaSi$_2$. We expect that these point defects form during the MBE growth when the $R_{Ba}/R_{Si}$ is deviated from the ideal value, and worked as donor impurities. Besides, the residual hole concentration seems to be $1 \times 10^{15}$ cm$^{-3}$. Therefore, the conduction type changes from n to p when $R_{Ba}/R_{Si} = 2.2$.