Possibility of Si-based new material for thin-film solar cell applications

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Abstract. We grow BaSi₂ epitaxial films on Si(111) substrates by molecular beam epitaxy, and investigate their optical properties such as optical absorption coefficients, minority-carrier diffusion length, and minority-carrier lifetime. These are key parameters which determine the solar cell performance. The band gap of BaSi₂ is measured to be approximately 1.3 eV. The absorption coefficient reaches approximately $3 \times 10^4$ cm⁻¹ at 1.5 eV. The minority-carrier diffusion length and minority-carrier lifetime are found to be about 10 µm and 8 µs, respectively. These values are sufficiently large for thin-film solar cell applications. Internal photoresponse spectra are deduced from data obtained experimentally.

1. Introduction
Recently photovoltaic solar cells have been attracting considerable attention. At present, approximately 90% of solar cells are based on crystalline Si because of its earth-abundance and developed technologies. Thin-film solar cell materials such as CIGS, CZTS, CdTe, and organic materials have been attracting increasing attention due to their high efficiency and low cost. In contrast, the optical absorption layers of crystalline Si solar cells tend to be much thicker than conventional thin-film solar cells, because the optical absorption coefficient $\alpha$ is much smaller for crystalline Si. Therefore, novel Si-based materials for high-efficiency thin-film solar cells have received significant interest. However, little steadfast effort has been devoted to any materials other than Si, CZTS, CIGS, CdTe and III-V compounds as far as inorganic semiconductors are concerned. Among such materials, we have focused much attention on semiconducting BaSi₂. The band gap of BaSi₂ is approximately 1.3 eV [1,2], and can be increased up to 1.4 eV in Ba₁₋ₓSrₓSi₂ [3], which matches the ideal solar spectrum much better than crystalline Si. In addition, BaSi₂ has a very large absorption coefficient of approximately $3 \times 10^5$ cm⁻¹ at 1.5 eV [2]. A large value of $\alpha$ and expansion of the band gap in Ba₁₋ₓSrₓSi₂ were theoretically expected [4,5]. In solar cells, both absorption coefficient $\alpha$ and minority-carrier diffusion length $L$ are critical. In order to efficiently extract photogenerated carriers, the diffusion length should be larger than the absorption length. Direct band gap semiconductors like GaAs usually have large absorption coefficient. But this generally comes with faster recombination and hence relatively smaller diffusion length. In contrast, both $\alpha$ and $L$ were found to be large enough in BaSi₂ ($L \sim 10$µm) [2,6]. This is because BaSi₂ is an indirect band gap semiconductor with a direct transition 0.1 eV above the band edge. Recently, we successfully achieved large photoresponsivity and internal quantum efficiency exceeding 70% in 0.4µm-thick $a$-axis-oriented BaSi₂ epitaxial layers [7].
Excess carrier recombination mechanisms in BaSi₂ have also been studied [8]. These results have spurred interest in this material.

Control of the conductivity of BaSi₂ by impurity doping is a requirement because the basic structure of a solar cell is a p-n junction. According to Imai and Watanabe [9], substitution of Si in the BaSi₂ lattice is more favorable than substitution of Ba from an energetic point of view by first-principles calculation. In our previous works, the electron concentration of Sb-doped n-type BaSi₂ was controlled in the range between 10¹⁶ and 10²⁰ cm⁻³ at room temperature (RT) [10]. Very recently, we have achieved the hole concentration exceeding 10¹⁹ cm⁻³ in B-doped p-type BaSi₂. The hole concentration of B-doped BaSi₂ was controlled in the range between 10¹⁶ and 10²⁰ cm⁻³ at RT. In this article, the current status and future prospect of this material towards high-efficiency and thin-film solar cell applications are reviewed.

2. Experimental procedures

An ultrahigh vacuum (UHV) chamber equipped with a Knudsen cell for Ba and an electron beam gun for Si was employed. Before the growth, the Si(111) substrates were prepared by subjecting them to the following treatment. The substrates were washed using RCA clean steps, which removed organic and metallic contaminants. The substrates were then annealed at 830 °C for 30 min in the UHV (1 × 10⁻⁶ Pa) chamber to remove the protective SiO₂ layers. After annealing, a 7×7 streaky reflection high-energy electron diffraction (RHEED) pattern was observed, indicating a clean Si surface. A two-step growth method was adopted, which included reactive deposition epitaxy (RDE; Ba deposition on hot Si) and molecular beam epitaxy (MBE; co-deposition of Ba and Si) to form thick BaSi₂ films [11,12]. The RDE process was carried out for deposition of a template layer as a BaSi₂ precursor prior to the subsequent MBE process. The same growth method was successfully utilized for the epitaxial growth of semiconducting β-FeSi₂ films on both Si(001) and Si(111) substrates [13,14]. The crystalline quality of the films was evaluated using RHEED and θ-2θ X-ray diffraction (XRD) measurements using Kα x-rays.

3. Results and discussions

3.1. Optical absorption properties

For measurements of optical absorption coefficients, we fabricated the silicon-on-insulator (SOI) substrates using wafer bonding at room temperature (RT) as well as chemical mechanical polishing (CMP). First, 500-µm-thick high-resistive FZ-Si(111) (ρ > 1000 Ω·cm) and 500-µm-thick fused silica wafers were bonded at RT. Then, the Si wafer was mechanically ground and polished by CMP down to about 0.7 µm thickness. Prior to loading the substrates into the MBE chamber, oxide layers on the surface of the SOI substrate were etched away by diluted hydrofluoric acid. Thermal treatment was performed at 590 °C for 20 min in UHV to clean the hydrogen-terminated surface. A 20-nm-thick BaSi₂ template layer was then formed at 525 °C using RDE, prior to the deposition of an 80-nm-thick BaSi₂ layer by MBE (sample A). Si and Ba atoms were co-deposited at 575°C by an electron-beam gun and a Knudsen cell, respectively. For comparison, BaSi₂ layers were also epitaxially grown on a Si(111) substrate (sample B) in the same manner as described previously [12]. RHEED and θ-2θ XRD patterns were utilized to evaluate the crystalline quality of the BaSi₂ layers. We employed a JASCO U-best 570 spectrophotometer to obtain the transmission spectra of the samples. We observed the (1×1) streaky pattern of Si(111) after thermal cleaning at 590°C.

Figures 1(a) and 1(b) show RHEED patterns of the RDE-grown BaSi₂ template layer. The incident electron beam was along the (a) Si[1-10] and (b) Si[11-2] directions. After the RDE growth, we confirmed that the BaSi₂ film was epitaxially grown on the Si layer of the SOI substrate as shown in Figs. 1(c) and 1(d). Since these patterns could also be observed at the end of MBE growth, we determined that the BaSi₂ film was indeed grown epitaxially.
Figure 2 shows the $\theta$-2$\theta$ XRD patterns of samples A and B, BaSi$_2$ films grown on the SOI and Si(111) substrates, respectively. We can see diffraction peaks only from (100)-oriented BaSi$_2$ planes, such as the (200), (400), (600) planes. For the BaSi$_2$ epitaxial film on the SOI substrate, the peak intensities of these planes were almost the same as those in the BaSi$_2$ epitaxial film on the Si(111) substrate. These results indicate that the crystalline quality of the BaSi$_2$ film on the SOI substrate was equivalent to that on the single-crystalline Si(111) substrate.

The transmission spectrum for the BaSi$_2$/SOI structure is presented in Fig. 3. This spectrum was significantly influenced by an interference effect within the 0.7-$\mu$m-thick Si layers. The Si layers were very thin and flat due to the CMP process, so that these interference fringes were superimposed on the spectrum. We used the equations presented in Ref. 15 to derive the interference-free transmission spectrum. As shown in Fig. 3, the transmission spectrum was fitted by the maximal extremes of the interference fringes ($T_M$) and also by their minimal extremes ($T_m$). Assuming that the Si layers have a certain absorption coefficient on a transparent substrate, the interference-free transmission spectrum $T_\alpha$ can be expressed as just the geometric mean of $T_M$ and $T_m$ ($T_\alpha=\sqrt{T_M \cdot T_m}$) over the entire region of the transmission spectrum. We calculated the $T_\alpha$ values using the $T_M$ and $T_m$ curves, and also eliminated the absorption due to the Si layers by measuring the transmission spectrum of another SOI substrate. We finally normalized $T_\alpha$ by taking the reflectivity of the sample into consideration. Using $T_\alpha$, we could obtain the absorption spectrum shown in Fig. 4(a). The absorption coefficient of BaSi$_2$ reached $3\times10^4$ cm$^{-1}$ at 1.5 eV. Figure 4(b) shows the $(\alpha d\nu)^{1/2}$ versus $\nu$ plot for deriving the indirect optical absorption edge. The straight fit line intersects the horizontal axis at 1.34 eV. Thus, the indirect absorption edge with phonon emission was 1.34 eV. The absorption with phonon absorption is neglected, because the fitting was performed at the higher-energy region than the indirect band gap. Thus, the indirect band gap is determined to be approximately 1.32 eV, assuming that the phonon energy is approximately 25 meV. It is true that absorption coefficients below $10^2$ cm$^{-1}$ are necessary to determine the precise energy gap. Much thicker BaSi$_2$ enables us to measure smaller absorption coefficients free from errors. In addition, we can’t rule out the possibility that defects contribute to absorption to some extent in the energy range below 1.34 eV.

**Figure 1.** RHEED patterns of BaSi$_2$ in sample A, observed after (a)(b) RDE and (c)(d)MBE growth along the (a)(c) Si[1-10] and (b)(d) Si[11-2] azimuths.

**Figure 2.** $\theta$-2$\theta$ XRD patterns for samples A and B, grown on SOI and Si(111) substrates, respectively. The forbidden diffraction peak of Si(222) indicated by asterisk occurs by double diffraction.
3.2. Minority-carrier properties

3.2.1. Minority-carrier diffusion length

For measurements of minority-carrier diffusion length in undoped BaSi$_2$, we fabricated 300-nm-thick undoped n-BaSi$_2$ films by RDE at 550 °C for 5 min, followed by MBE at 600 °C for 120 min. Undoped BaSi$_2$ shows n-type conductivity with electron concentrations of approximately $10^{16}$ cm$^{-3}$ [1].

In order to investigate the grain size of BaSi$_2$ and grain boundaries (GBs), plan-view transmission electron microscopy (TEM) samples prepared by mechanical polishing and ion milling were observed using TOPCON EM-002B operated at 120 kV. For electron beam induced current (EBIC) measurements, Al/n-BaSi$_2$ Schottky diodes were formed. Front-side Schottky contacts were formed with Al on the BaSi$_2$ surface via wire bonding, and the back-side ohmic contact was made with Al by sputtering. EBIC observations were carried out in the edge-scan configuration with a Hitachi S4300 field-emission scanning electron microscope (SEM) in the EBIC mode at RT. The acceleration voltage of the electron beam, $V_{ac}$, was set at 5 kV to avoid penetration of the beam into the Si substrate. The penetration depth of the electron beam is estimated to be shorter than 300 nm, which is the thickness of the BaSi$_2$ layers, when $V_{ac}$ is 5 kV, with the density of BaSi$_2$ being 5.14 g/cm$^3$.

Figures 5(a)(b) and 5(c)(d) show secondary-electron (SE) and EBIC images around the Al contact, respectively, with $V_{ac}$ = 5 kV. In the EBIC method, carriers generated within the diffusion length in the n-type BaSi$_2$ are collected by the electric field under the Al contact and sensed as a current in the external circuit. In Figs. 5(c)(d), the brighter regions show higher collection of electron-beam-induced carriers in the BaSi$_2$. We cannot see defect-related black lines. Figure 6 shows the EBIC line-scan data along dotted line AA’ in Fig. 5(c). The EBIC profile shows an exponential dependence of the distance from the Al contact. In this work, the diffusion length of minority carriers was roughly estimated to be approximately 10 $\mu$m, assuming that the EBIC profile varies as $\exp(-x/L)$, where $x$ is the distance from the Al edge (point A) along the dotted line, and $L$ is the diffusion length of holes for BaSi$_2$. The obtained minority-carrier diffusion length is much larger than the grain size of the BaSi$_2$, implying that the GBs do not work as defect centers for minority carriers in n-BaSi$_2$. The contribution of carriers generated within the n-Si substrate to the measured EBIC signals can be excluded, because the simulated penetration depth of the electron beam is shorter than 300 nm, the thickness of the BaSi$_2$ layers, when $V_{ac}$ is 5 kV. Thus, it is reasonable to think that the number of carriers generated within the Si substrate was negligibly small compared to those generated in the BaSi$_2$ layers. To confirm the GBs character, an EBIC system with spatial resolutions less than 0.1 $\mu$m may be necessary. We should also
note here that the obtained minority-carrier diffusion length is roughly 30 times larger than $1/\alpha = (0.3 \mu m)$ at 1.5 eV, suggesting that this value is large enough for solar cell applications.

**Figure 5.** (a)(b) SE and (b)(d) EBIC images around the Al contact.

3.2.2. Minority-carrier lifetime diffusion length

After cleaning a floating-zone n-Si(111) substrate ($\rho > 1000 \Omega \cdot cm$) at 900 °C for 30 min in UHV, a 5-nm-thick BaSi$_2$ template layer was grown on the Si substrate by RDE, followed by MBE to form a 1.5-μm-thick BaSi$_2$ epitaxial film. Figure 7 shows the relationship between apparent minority-carrier life time $\tau_a$ and excess-carrier density of the 1.5-μm-thick BaSi$_2$. The undoped BaSi$_2$ shows n-type conductivity and the electron concentration is about $10^{16} \text{ cm}^{-3}$ [1]. The excess carrier concentration was calculated from the absorption coefficient of BaSi$_2$ at 349 nm, and the irradiated laser intensity, which was varied from $1.1 \times 10^2$ to $1.3 \times 10^2 \text{ W/cm}^2$. The inset shows the decay curve when the excess carrier concentration was $7.4 \times 10^{16} \text{ cm}^{-3}$. Decay can be divided into two modes in terms of decay rate. We see initial rapid decay, followed by approximately constant decay. According to our previous work, the rapid decay is caused by Auger recombination [8]. $\tau_a$ was calculated by approximating the tail region of the decay curve by a single exponential curve. We see that $\tau_a$ decreases with increasing the excess-carrier density. This is because the excess-carrier density is several orders of magnitudes higher than majority-carrier density at equilibrium in undoped n-BaSi$_2$. Such high carrier injection leads to multicarrier recombination. Therefore, when we discuss about the minority-carrier lifetime, we should use lower injection value. As can be seen in Fig. 7, $\tau_a$ becomes independent of excess-carrier density when the excess carrier concentration becomes smaller than $10^{16} \text{ cm}^{-3}$, indicating that $\tau_a$ corresponds to the effective minority-carrier lifetime $\tau$ in this region. This result makes sense because the electron concentration of undoped n-BaSi$_2$ is about $10^{16} \text{ cm}^{-3}$ [1]. Therefore, it

**Figure 6.** Experimental and simulated (solid line) EBIC line-scan profiles along the dotted line from points A to A’ in Fig. 5(c).

**Figure 7.** Relationship between minority-carrier life time and excess-carrier density of 1.5-μm-thick BaSi$_2$. The inset shows the photoconductivity decay curve of the 1.5-μm-thick BaSi$_2$ film when the excess-carrier concentration is $7.4 \times 10^{16} \text{ cm}^{-3}$. 

is reasonable to think that the minority-carrier lifetime in the 1.5-µm-thick BaSi$_2$ is about 8 µs. Assuming that the hole mobility is 1 cm$^2$/V·s$^{-1}$ [16], the minority-carrier diffusion length $L$ is estimated to be about 4.5 µm from the equation of $L = \sqrt{\frac{k_B T \mu \tau}{q}}$. Here, $k_B$ is the Boltzmann constant, $T$ the absolute temperature, $\mu$ the mobility of holes, $\tau$ the effective minority-carrier lifetime, and $q$ the elemental charge. This value is about three times larger than the absorption layer thickness (~1.5 µm) in a BaSi$_2$ pn junction diode. On the basis of these results, we achieved the formation of high-quality BaSi$_2$ films thicker than 1.5 µm, where the minority-carrier lifetime was large enough for thin-film solar cell applications.

3.3. Expected spectral response in a BaSi$_2$ pn junction diode

We attempted to deduce the expected photocurrent density in a BaSi$_2$ p’n homojunction diode. Analytical expressions for the photogenerated and dark saturation current densities can be obtained only when the dopant concentration, carrier lifetime and other parameters are assumed constant. Suppose here a simple p’n abrupt junction diode in samples pn-A to pn-E as shown in Table 1. $N_A$, $W_e$, $L_e$, $S_e$ are the acceptor density, neutral p+ layer width, minority-carrier (electrons) diffusion length, and surface recombination velocity, and $N_D$, $W_b$, $L_h$, and $S_h$ are their counterparts in the n layer. $W$ is the depletion region width.

Table 1. Parameters of BaSi$_2$ homojunction diode are shown.

| Sample | $N_A$ (cm$^{-3}$) | $W_e$ (µm) | $L_e$ (µm) | $S_e$ (cm/s) | $W$ (µm) | $N_D$ (cm$^{-3}$) | $W_b$ (µm) | $L_h$ (µm) | $S_h$ (cm/s) |
|--------|------------------|------------|------------|-------------|----------|------------------|------------|------------|-------------|
| pn-A   | $10^{18}$        | 0.1        | 0.1        | 8           | 0.4      | $10^{16}$        | 1.5        | 10         | 8           |
| pn-B   | $10^{18}$        | 0.1        | 0.1        | 8           | 0.4      | $10^{16}$        | 2.5        | 10         | 8           |
| pn-C   | $10^{18}$        | 0.05       | 0.1        | 8           | 0.4      | $10^{16}$        | 1.5        | 10         | 8           |
| pn-D   | $10^{18}$        | 0.05       | 0.1        | 100         | 0.4      | $10^{16}$        | 1.5        | 10         | 8           |
| pn-E   | $10^{18}$        | 0.05       | 0.1        | 8           | 0.4      | $10^{16}$        | 1.5        | 10         | 100         |

The total photocurrent density $j$ at energy $E$ per unit energy is given by the sum of current due to electrons generated within the neutral p$^+$-BaSi$_2$ region, $j_e$, current due to electron-hole pairs generated within the depletion region, $j_{DL}$, and current due to holes generated within the neutral n-BaSi$_2$ region, $j_h$. The internal quantum efficiency, $IQE$, is equal to,

$$IQE(E) = \frac{j}{q \Phi_0 (1-R)} = \frac{j_e + j_{DL} + j_h}{q \Phi_0 (1-R)}$$

(1)

Once the $IQE$ is derived, the $J_L$ is given by,

$$J_L = q \int \Phi_0 (1-R) IQE(E) dE$$

(2)

In order to make $J_L$ large, we should minimize $R$ and maximize $IQE$ over the solar spectral range. Therefore, the $IQE$ is a measure of the quality of the diode. The contribution to the $IQE$ by $j_{DL}$, $j_e$ and $j_h$, that is $IQE_{DL}$, $IQE_e$, and $IQE_h$, respectively, are obtained analytically. Then, we investigated the dependence of $IQE$ on various parameters. Regarding the $\alpha$, we used the experimental data shown in ref 1. While the diffusivity is not highly sensitive to impurity and defect, we thus set both $D_e$ and $D_h$ to be 0.1 cm$^2$/s, because $L_h$ is 10 µm and $\tau_h$ is 10 µs as described earlier. When the impurity concentrations in the p$^+$ and n layers, that is $N_A$ and $N_D$, are set to be $10^{18}$ and $10^{16}$ cm$^{-3}$, respectively. In this case, the $W$ is estimated to be 0.4 µm and the built-in voltage 1.0 V assuming that the permittivity of BaSi$_2$ is approximately 15, and the effective densities of states of the conduction band.
and valence band, that is, \( N_C \) and \( N_V \), respectively, are \( 2.6 \times 10^{19} \) and \( 2.0 \times 10^{19} \) cm\(^{-3}\). The square of the intrinsic carrier concentration, \( n_i \), at RT is calculated to be \( n_i^2 = 8 \times 10^{16} \) cm\(^{-3}\). Other parameters are summarized in Table 1. In Figs. 8(a) and 1(b), the IQE is almost the same between samples pn-A and pn-B although the \( W_b \) is increased from 1.5 \( \mu \)m in sample pn-A to 2.5 \( \mu \)m in sample pn-B, meaning that \( W_b=1.5 \) \( \mu \)m is enough. We see that when the \( W_e \) is decreased from 0.1 to 0.05 \( \mu \)m, the IQE is increased particularly in higher energies in samples pn-C to pn-E. This is attributed to the enhancement of IQE\(_e\) and IQE\(_{DL}\). Since the absorption coefficient of BaSi\(_2\) is large, the contributions of the surface p’-layer and the depletion region to the IQE becomes pronounced when the \( W_c \) is decreased. This result implies that we design the \( W_b \) to be equal to \( L_e \) or smaller. In samples pn-C to pn-E, there is no significant difference in IQE regardless of different \( S_e \) and \( S_h \). This means that the IQE is not so sensitive to surface conditions in a BaSi\(_2\) homojunction solar cell. We next discuss the expected conversion efficiency of the diodes. Considering that \( n_{p0} \) is much smaller than \( p_{n0} \) because of the p’n junction, the dark current density of the diode, \( J_{dark} \), is dominant from the contribution of n-type layer, and thus, the reverse saturation current density \( J_s \) is obtain. The \( J_s \) and \( J_L \) values, and the expected open-circuit voltage, \( V_{OC} \), under AM1.5 illumination for samples pn-A to pn-E are investigated, assuming that the fill-factor, \( FF \), is 0.8 regardless of \( V_{OC} \) and \( R=0 \). As a result, the conversion efficiency is calculated to be more than 25\% in samples pn-C and pn-D even though the total layer thickness is only 2 \( \mu \)m.

**Figure 8.** Calculated internal spectral responses for samples pn-A to pn-E. Individual contributions of IQE\(_e\), IQE\(_{DL}\), and IQE\(_h\) to IQE are shown.
4. Summary

Current status toward Si-based high-efficiency thin-film solar cells was reviewed in terms of optical absorption properties and minority-carrier properties. These are key parameters which determine the performance of solar cells. The band gap of BaSi$_2$ was measured to be approximately 1.3 eV. The absorption coefficient reached approximately $3 \times 10^4$ cm$^{-1}$ at 1.5 eV. The minority-carrier diffusion length and minority-carrier lifetime were found to be about 10 µm and 8 µs, respectively. In particular, what’s interesting with BaSi$_2$ is that it has large absorption coefficients and at the same time large minority-carrier diffusion length. This feature, caused by the fact that the direct transition starts just 0.1 eV above the band edge, is favourable to effectively extract photogenerated carriers. We calculated the photoresponse spectra of BaSi$_2$ p+n homojunction diodes using experimentally obtained absorption coefficients and minority-carrier diffusion length, and discussed the influence of parameters such as emitter and base layer thicknesses, surface recombination velocities. The IQE is not so sensitive to $S_e$ and $S_h$, and $W_b$, while the $W_e$ has a significant influence on IQE particularly in a high energy region. This is because the absorption coefficient of BaSi$_2$ is large. Thus, we need to take care of the relationship between $W_e$ and $L_e$ in the emitter layer when we design the $W_e$. The conversion efficiency exceeding 25% is expected for 2 µm-thick BaSi$_2$ homojunction diodes in an ideal condition.

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