

# Decrease in electrical contact resistance of Sb-doped n+-BaSi<sub>2</sub> layers and spectral response of an Sb-doped n+-BaSi<sub>2</sub>/undoped BaSi<sub>2</sub> structure for solar cells

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# **Decrease in electrical contact resistance of Sb-doped n<sup>+</sup>-BaSi<sub>2</sub> layers and spectral response of an Sb-doped n<sup>+</sup>-BaSi<sub>2</sub>/undoped BaSi<sub>2</sub> structure for solar cells**

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We investigated how the electron concentration  $n$  in a 300-nm-thick Sb-doped n<sup>+</sup>-BaSi<sub>2</sub> layer grown by molecular beam epitaxy affected the contact resistance  $R_C$  to surface electrodes (Al, indium-tin-oxide). As the  $n$  of n-BaSi<sub>2</sub> increased,  $R_C$  decreased and reached a minimum of 0.019  $\Omega$  cm<sup>2</sup> at  $n = 2.4 \times 10^{18}$  cm<sup>-3</sup> for the Al electrodes. This value was more than one order of magnitude smaller than that obtained for Al/B-doped p-BaSi<sub>2</sub>. We believe that this significant decrease in  $R_C$  came from Sb segregation. Furthermore, the internal quantum efficiency (*IQE*) spectrum was evaluated for an Sb-doped n<sup>+</sup>-BaSi<sub>2</sub>(20 nm)/undoped BaSi<sub>2</sub>(500 nm)/n<sup>+</sup>-Si(111) structure. Its *IQE* reached as high as ~50% over a wide wavelength range under a small bias voltage of 0.1 V applied between the top and bottom electrodes.

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## 1. Introduction

In many studies, various solar cells materials such as CdTe, chalcopyrites, kesterite, and perovskite have been assessed because of their low cost and high efficiency [1-6]. However, these materials contain elements that are scarce, toxic, or both. Thin-film Si solar cells have also been studied extensively; however, it is not easy to achieve efficiencies as high as 20% [7-17]. Thus, it is important to explore other materials for thin-film solar cells. Among such materials, we have focused on barium disilicide ( $\text{BaSi}_2$ ), a semiconductor consisting of only abundant elements [18,19].  $\text{BaSi}_2$  is attractive for use in thin-film solar cells because of its bandgap of 1.3 eV, a matching solar spectrum [20], a high absorption coefficient of  $3 \times 10^4 \text{ cm}^{-1}$  at 1.5 eV comparable to those of chalcopyrites [21-23], and superior minority-carrier diffusion length ( $L \sim 10 \text{ }\mu\text{m}$ ) [24,25] and lifetime ( $\tau \sim 10 \text{ }\mu\text{s}$ ) [26-29]. Because of these properties, an energy conversion efficiency  $\eta$  of over 25% can be expected for a 2- $\mu\text{m}$ -thick  $\text{BaSi}_2$  pn-junction diode [30]. Recently, we have achieved a conversion efficiency near 10% with B-doped p- $\text{BaSi}_2$ /n-Si heterojunction solar cells [31].

To fabricate  $\text{BaSi}_2$  homojunction solar cells, we plan to use undoped  $\text{BaSi}_2$  as an absorber layer in a solar cell. Undoped  $\text{BaSi}_2$  has both a high  $\alpha$  and a large  $L$ , both of which facilitate the collection of photogenerated carriers in an external circuit even though  $L$  degrades somewhat because of extrinsic effects such as crystal imperfections. In a solar cell, the topmost layer should be a heavily impurity-doped n- or p-type layer to decrease the contact resistance  $R_C$ , which directly affects the fill factor  $FF$  of the solar cell. As  $R_C$  increases,  $FF$  decreases and thus  $\eta$  decreases. In many studies,  $R_C$  have been assessed in solar cell materials [32-38]. In our previous work, we investigated the  $R_C$  between Al electrodes and B-doped p- $\text{BaSi}_2$ . The  $R_C$  reached a minimum of  $0.35 \text{ }\Omega \text{ cm}^2$  [31] when the hole concentration  $p$  of p- $\text{BaSi}_2$  was  $4.0 \times 10^{18} \text{ cm}^{-3}$ . In this study, we first investigated the dependence of  $R_C$  between surface electrodes (Al or ITO) and Sb-doped n- $\text{BaSi}_2$  layers at various electron concentrations. We then evaluated the

spectral response of an Sb-doped  $n^+$ -BaSi<sub>2</sub>(20 nm)/undoped BaSi<sub>2</sub>(500 nm) structure to verify how the heavily doped  $n^+$ -BaSi<sub>2</sub> surface layer affected the spectral response, especially at short wavelengths.

## 2. Experimental methods

Here, we used an ion-pumped molecular beam epitaxy (MBE) system equipped with an electron-beam evaporation source for Si as well as standard Knudsen cells for Ba and Sb. Details of the growth procedure of undoped BaSi<sub>2</sub> and Sb-doped  $n$ -BaSi<sub>2</sub> were reported previously [39,40]. To measure  $R_C$ , we used high-resistivity Czochralski (CZ) p-Si(111) substrates (resistivity  $\rho = 1000\text{--}10000 \text{ } \Omega \text{ cm}$ ) to minimize current flowing through the substrate. After thermally cleaning the substrate at the substrate temperature  $T_S = 900 \text{ } ^\circ\text{C}$ , we deposited Ba at  $500 \text{ } ^\circ\text{C}$  to form an approximately 5-nm-thick BaSi<sub>2</sub> template layer by reactive deposition epitaxy. This template worked as seeds that control the crystal orientation of the BaSi<sub>2</sub> overlayers [41]. Next, we formed 300-nm-thick Sb-doped  $n$ -BaSi<sub>2</sub> epitaxial layers by MBE at various  $T_S$  values of 460 (sample A), 480 (sample B), 500 (sample C), and  $520 \text{ } ^\circ\text{C}$  (sample D). The electron concentration  $n$  of Sb-doped  $n$ -BaSi<sub>2</sub> depends on  $T_S$  [40]. Then, a 3-nm-thick a-Si passivation layer was deposited *in situ* at  $T_S = 180 \text{ } ^\circ\text{C}$  [42]. Finally, 150-nm-thick striped Al or ITO electrodes (length  $l = 0.6 \text{ mm}$ , width  $W = 7.0 \text{ mm}$ , electrode interval  $d = 1.0 \text{ mm}$ ) were formed by sputtering. Carrier concentration was measured by the van der Pauw method at room temperature (RT).  $R_C$  was measured by the transfer length method at RT [43]. We compared the smallest  $R_C$ ,  $R_C = 0.35 \text{ } \Omega \text{ cm}^2$ , measured from Al/p-BaSi<sub>2</sub> ( $p = 4.0 \times 10^{18} \text{ cm}^{-3}$ , sample E) [31]. The depth profile of Sb atoms was measured by secondary ion mass spectrometry (SIMS) using Cs<sup>+</sup> ions.

Next, we investigated the photoresponse properties of the Sb-doped  $n^+$ -BaSi<sub>2</sub>/undoped BaSi<sub>2</sub> structure (sample F). A 500-nm-thick undoped BaSi<sub>2</sub> layer was epitaxially grown on a

low-resistivity Cz n-Si(111) substrate ( $\rho < 0.01 \text{ } \Omega \text{ cm}$ ) at  $T_S = 580 \text{ } ^\circ\text{C}$ , followed by a 20-nm-thick Sb-doped n<sup>+</sup>-BaSi<sub>2</sub> ( $n = 2.4 \times 10^{18} \text{ cm}^{-3}$ ) layer and a 3-nm-thick a-Si passivation layer. We used the low-resistivity Si substrate to exclude the contribution of photogenerated carriers in the n-Si substrate and to form a good electrical contact at the BaSi<sub>2</sub>/Si heterointerface. Finally, 80-nm-thick ITO electrodes with a diameter of 1 mm and 150-nm-thick Al rear electrodes were formed by sputtering. The reflectance  $R$  and external quantum efficiency ( $EQE$ ) spectra were evaluated at RT using a lock-in technique with a xenon lamp and a single monochromator (Bunko Keiki, SM-1700A and RU-60N) with a focal length of 25 cm. Internal quantum efficiency ( $IQE$ ) was obtained by dividing the  $EQE$  by  $(1-R)$ . Light intensity was calibrated using a pyroelectric sensor (Melles Griot, 13PEM001/J). The crystalline quality of the grown layers was characterized by reflection high-energy electron diffraction (RHEED) and X-ray diffraction (XRD) analyses using Cu K $\alpha$  radiation.

### 3. Results and discussion

Figure 1 shows the  $\theta$ - $2\theta$  XRD and RHEED patterns of samples A, B, C, and D. The RHEED patterns were observed along the Si  $[1\bar{1}2]$  azimuth. The diffraction peaks appeared only from the (100)-oriented BaSi<sub>2</sub> planes, such as (200), (400), and (600), showing that  $a$ -axis-oriented BaSi<sub>2</sub> epitaxial films were formed. However, the RHEED patterns were not streaky but spotty, meaning that the surface is rough. This surface roughness developed because the  $T_S$  for Sb-doped BaSi<sub>2</sub> was much lower [40] than the optimum  $T_S$  ( $\sim 600 \text{ } ^\circ\text{C}$ ) for the undoped BaSi<sub>2</sub> absorber layers. The  $n$  values of samples A, B, C, and D were  $6.9 \times 10^{17}$ ,  $1.8 \times 10^{18}$ ,  $2.4 \times 10^{18}$ , and  $2.8 \times 10^{17} \text{ cm}^{-3}$ , respectively. Now, using sample C as an example, we will show how to obtain  $R_C$ . Figure 2 shows the relationship between the measured resistance  $r(x)$  between the Al or ITO electrodes and the electrode distance  $x$  for sample C ( $n = 2.4 \times 10^{18} \text{ cm}^{-3}$ ,  $\mu = 100 \text{ cm}^2/\text{V s}$ ). Ignoring the lateral resistance of the electrode when the current flows in the  $x$  direction,

$r(x)$  can be written as

$$r(x) \approx 2r_c + r_{\text{semi}}(x), \quad (1)$$

where  $r_{\text{semi}}(x)$  is the resistance of the semiconductor and  $2r_c$  is the intercept of the vertical axis when we plot  $r(x)$  with respect to  $x$ . Because the second term on the right side of Eq. (1) is proportional to  $x$ ,  $r(x)$  increases with increasing  $x$ . As shown in Fig. 2, the line for the ITO electrodes has a slightly higher slope and thus a larger resistance than that for Al electrodes. This behavior probably occurred because the ITO electrode had larger lateral resistance than Al. From the plots in Fig. 2,  $r_c = 2.26$  and  $1.22 \text{ } \Omega$  were obtained for the ITO and Al electrodes, respectively.  $R_C$  is given by

$$R_C = r_c \times W \times L_t, \quad (2)$$

where  $L_t$  is the transfer length, which is half of the intercept of the horizontal axis of the straight line in Fig. 2. From Eq. (2),  $R_C$  was calculated to be  $0.057$  and  $0.019 \text{ } \Omega \text{ cm}^2$  for the ITO and Al electrodes, respectively, in sample C.

Figure 3 summarizes the dependence of  $R_C$  on  $n$  for all the samples obtained by the method described above.  $R_C$  decreased with increasing  $n$ . The smallest  $R_C$ ,  $0.019 \text{ } \Omega \cdot \text{cm}^2$ , was obtained from Al electrodes in sample C. This value is more than one order of magnitude smaller than the smallest  $R_C$  ( $0.35 \text{ } \Omega \text{ cm}^2$ ) obtained from Al/B-doped p-BaSi<sub>2</sub> ( $p = 4.0 \times 10^{18} \text{ cm}^{-3}$ ,  $\mu = 28 \text{ cm}^2 \text{V}^{-1} \text{ s}^{-1}$ ) [31]. We will now discuss the reason for this behavior. According to Ref. 36, the sheet resistance  $R_{\text{sheet}}$  is given by

$$R_{\text{sheet}} = R_C / L_t^2. \quad (2)$$

For sample C,  $R_{\text{sheet}}$  is  $0.40 \text{ } \Omega$  from  $L_t = 0.218 \text{ mm}$  and  $R_C = 0.019 \text{ } \Omega \text{ cm}^2$ , while  $R_{\text{sheet}}$  equals  $6.79 \text{ } \Omega$  using  $L_t = 0.227 \text{ mm}$  and  $R_C = 0.35 \text{ } \Omega \cdot \text{cm}^2$  for sample E. Such a large difference in  $R_{\text{sheet}}$  between samples C and E cannot be anticipated from their similar  $\rho$  values:  $0.0644$  and  $0.0557 \text{ } \Omega \text{ cm}$ , respectively, which were determined by Hall measurements. It is known that B atoms in B-doped p-BaSi<sub>2</sub> do not segregate and so are almost uniformly distributed [44]. Figure 4 shows

the depth profile of Sb atoms and secondary ions (Ba + Si) in sample C. The Sb atoms segregate toward the surface of sample C, and the Sb concentration exceeds  $10^{20} \text{ cm}^{-3}$  near the electrode. Segregation of Sb has been observed in various semiconductors such as Si, Ge, and III–V semiconductors [45-52]. Hence,  $n$  is actually much higher in the surface region than the  $n$  found from the Hall measurement ( $n = 2.4 \times 10^{18} \text{ cm}^{-3}$ ), leading to the significant decrease in  $R_{\text{sheet}}$  and thus  $R_c$  in sample C. Owing to the segregation of Sb in BaSi<sub>2</sub>, an Sb-doped n<sup>+</sup>-BaSi<sub>2</sub> layer should be placed on the top of the BaSi<sub>2</sub> solar cell. Hence, an Sb-doped n<sup>+</sup>-BaSi<sub>2</sub>/undoped BaSi<sub>2</sub> absorber layered structure is useful for practical applications.

We next assessed the spectral response of sample F, with an Sb-doped n<sup>+</sup>-BaSi<sub>2</sub>/undoped BaSi<sub>2</sub> structure. Figure 5 shows a schematic of sample F for this purpose and the RHEED patterns obtained after the growth of each layer. Because BaSi<sub>2</sub> has a large  $\alpha$ , short-wavelength light is absorbed near the surface, in the Sb-doped n<sup>+</sup>-BaSi<sub>2</sub> layer. Therefore, defective n<sup>+</sup>-BaSi<sub>2</sub> deteriorates the solar-cell performance. Figure 6 shows the  $\theta$ - $2\theta$  XRD pattern, revealing  $a$ -axis-oriented epitaxial growth up to the 20-nm-thick Sb-doped n<sup>+</sup>-BaSi<sub>2</sub>. Figure 7 shows the *IQE* spectrum of sample F. A bias voltage of  $-0.1 \text{ V}$  was applied to the top ITO electrode with respect to the back side-surface Al electrode so that the photogenerated holes in the BaSi<sub>2</sub> were extracted into the ITO electrode. The *IQE* increased sharply at wavelengths shorter than approximately 1000 nm, corresponding to the band gap of BaSi<sub>2</sub>, and reached approximately 50% over a wide wavelength range even under this small bias voltage. We observed no pronounced reduction in *IQE* at short wavelengths, even when a highly Sb-doped layer was inserted. In a BaSi<sub>2</sub> homojunction solar cell, the built-in potential across the pn junction will be much higher than 0.1 V, producing larger *IQEs*. On the basis of these findings, we conclude that the Sb-doped n<sup>+</sup>-BaSi<sub>2</sub>/undoped BaSi<sub>2</sub> absorber layered structure is suitable for practical BaSi<sub>2</sub> solar cells.

#### 4. Conclusions

We investigated the dependence of  $R_C$  on the electron concentration of Sb-doped n-BaSi<sub>2</sub> and achieved a minimum  $R_C$  of 0.019  $\Omega \text{ cm}^2$  for Al/Sb-doped n-BaSi<sub>2</sub> at  $n = 2.4 \times 10^{18} \text{ cm}^{-3}$ . This  $R_C$  was more than one order of magnitude smaller than the  $R_C$  obtained for Al/B-doped p-BaSi<sub>2</sub>, 0.35  $\Omega \cdot \text{cm}^2$ . We explained this significant reduction in  $R_C$  in Al/Sb-doped n-BaSi<sub>2</sub> as being due to Sb segregation, which was verified by SIMS. The concentration of Sb atoms exceeded  $10^{20} \text{ cm}^{-3}$  near the electrode, significantly decreasing  $R_C$ . We achieved an  $IQE > 50\%$  over a wide wavelength range under a small bias voltage of 0.1 V in the n<sup>+</sup>(20 nm)/undoped BaSi<sub>2</sub>(500 nm)/n<sup>+</sup>-Si(111) structure.

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## Figure captions

Fig. 1  $\theta$ - $2\theta$  XRD and RHEED patterns observed along the Si  $[1\bar{1}\bar{2}]$  azimuth of samples A, B, C, and D. The asterisk (\*) indicates the diffraction of the Si substrate used.

Fig. 2 Sample structure for  $R_C$  measurement and the relationships between resistance and Al or ITO electrode distance for sample C, Sb-doped n-BaSi<sub>2</sub> ( $n = 2.4 \times 10^{18} \text{ cm}^{-3}$ ).

Fig. 3  $R_C$  as a function of electron concentration for Al/n-BaSi<sub>2</sub> and ITO/n-BaSi<sub>2</sub>. The smallest  $R_C$  ( $0.35 \Omega \text{ cm}^2$ ) obtained from Al/p-BaSi<sub>2</sub> [31] is shown for comparison.

Fig. 4 SIMS depth profile of Sb atoms and secondary ions (Ba + Si).

Fig. 5 Sample structure of sample F and RHEED patterns of each layer observed along the Si  $[\bar{1}\bar{1}0]$  azimuth.

Fig. 6  $\theta$ - $2\theta$  XRD pattern of sample F. The asterisk (\*) indicates the diffraction of the Si substrate used.

Fig. 7  $IQE$  spectrum of sample F under a bias voltage of  $-0.1 \text{ V}$  applied to the surface ITO electrode with respect to the back side-surface Al electrode.

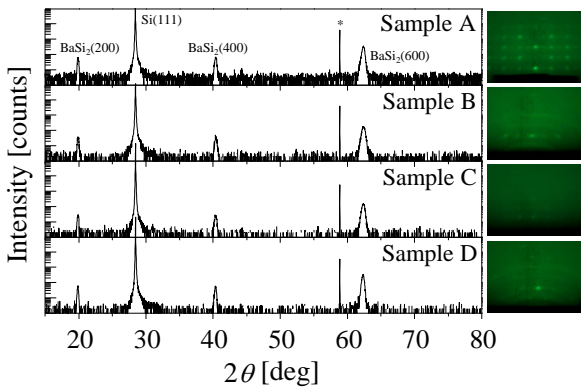


Fig. 1

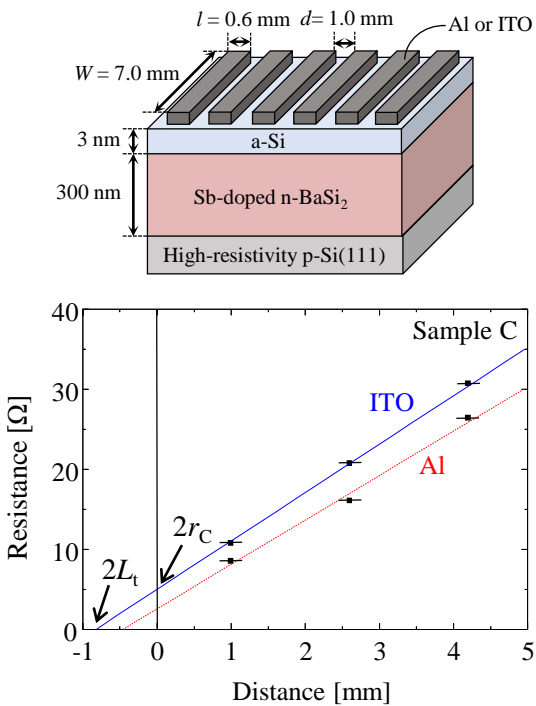


Fig. 2

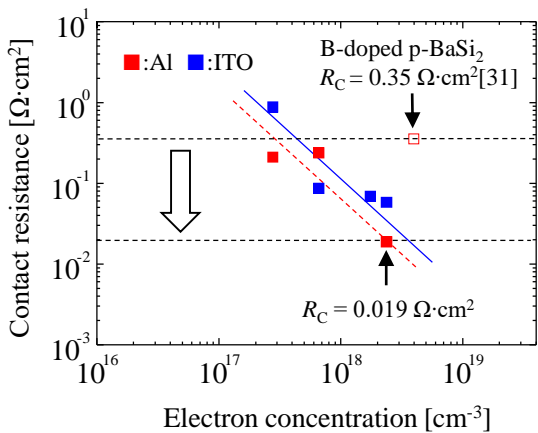


Fig. 3



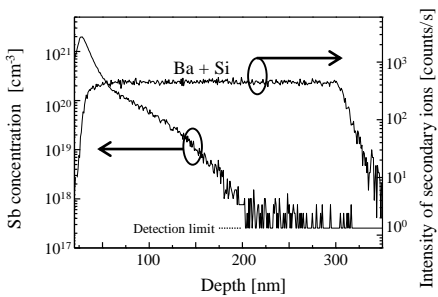


Fig. 4

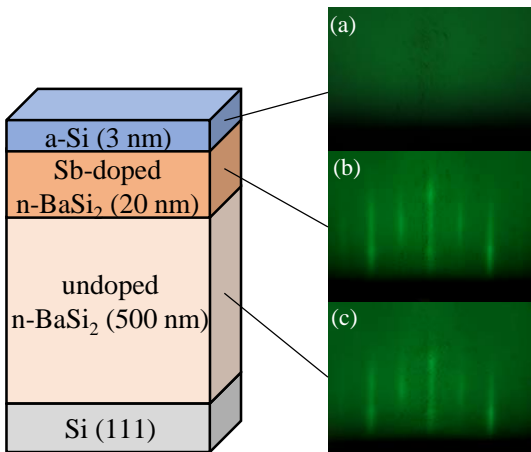


Fig. 5

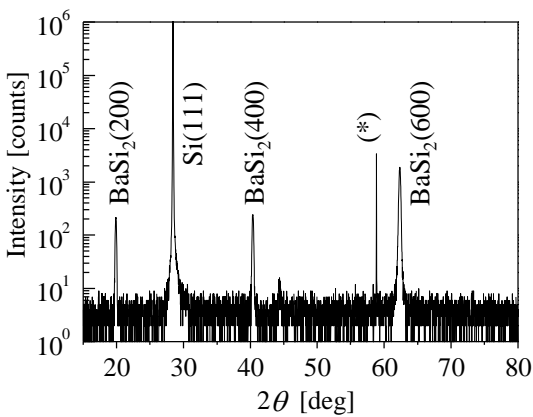


Fig. 6

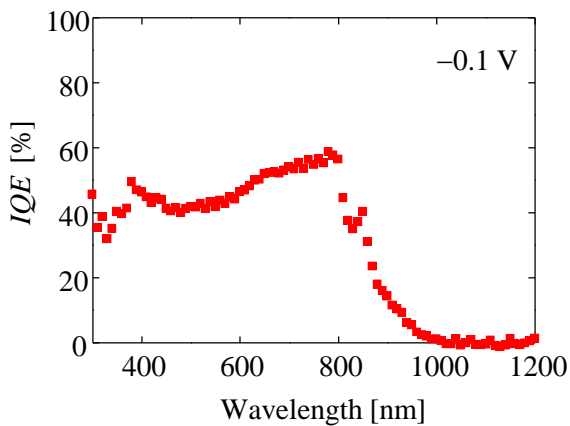


Fig. 7