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β -FeSi₂: A high refractive index candidate material for infrared bandpass filters

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ABSTRACT

Bandpass filters (BPFs) are optical filters with significantly high transmittance in a specific wavelength range and low transmittance on both sides. Infrared BPFs can reduce system losses and overheating caused by other light wavelengths owing to their ability to selectively transmit infrared light of the desired wavelength. This article discusses the potential of using a high refractive index material, β -FeSi₂, in BPFs. To the best of our knowledge, no studies have applied β -FeSi₂ to infrared BPFs. Simulation results showed that its high refractive index allows the excellent performance of the BPF to be achieved using a multilayer thin film structure with only three layers. Fourier transform infrared spectroscopy and x-ray photoelectron spectroscopy results showed that the β -FeSi₂ thin film exhibited the lowest absorptance of approximately 0 when the correct stoichiometry (Fe:Si = 1:2) was achieved through co-sputtering. Based on these findings, a β -FeSi₂/SiO₂/ β -FeSi₂ multilayer thin film was designed to fabricate the BPF. The fabricated BPF exhibited a narrow peak and achieved a peak transmittance exceeding 80%. This suggested that β -FeSi₂ is a promising material for fabricating infrared BPFs. Utilizing these filters is expected to yield significant efficiency improvements and reduce losses across various applications, including thermophotovoltaics and infrared heaters.

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I. INTRODUCTION

Bandpass filters (BPFs), also known as wavelength-selective transmission thin films, are optical filters with significantly high transmittance in a specific wavelength range and low transmittance on both sides. Owing to its ability to selectively transmit infrared light of the desired wavelength, an infrared BPF can reduce system losses and overheating caused by other light wavelengths. This characteristic has generated significant interest in employing infrared BPFs to enhance the power generation efficiency of thermophotovoltaic (TPV) systems, which are expected to break the Shockley–Queisser limit.^{1–4} Furthermore, infrared BPFs are critical in developing infrared heaters^{5,6} and gas sensor devices.⁷

Numerous studies have focused on achieving an ideal BPF with narrow and high peaks at the required wavelengths. Multilayer thin films composed of high and low refractive index dielectric materials that are transparent in the infrared region and arranged in a layer-by-layer structure have been reported to exhibit suitable transmittance spectra. However, the refractive index of materials commonly used in multilayer thin films, such as Si and TiO₂,^{8,9} is not sufficiently high, typically in the order of 2–3. In such cases, an increase in the thickness of the thin film and the number of layers

is required to achieve ideal optical functions. Other studies have suggested that two-dimensional¹⁰ and three-dimensional photonic crystals¹¹ can effectively control the shape and position of the peak by precisely tuning the wavelength of the photonic bandgap using their periodic structures. However, fabrication and stability challenges at high temperatures pose significant hurdles to this approach. Therefore, a new high refractive index material for infrared BPF is required to overcome these challenges.

β -FeSi₂ is well known for its remarkable optical and electrical properties. It has a direct bandgap of 0.87 eV and a significant optical absorption coefficient ($\alpha > 10^5$ cm⁻¹). Thus, β -FeSi₂ is a promising candidate for advanced optical applications, such as light-emitting diodes¹² and solar cells.¹³ Meanwhile, because of its high heat and oxidation resistance, β -FeSi₂ has been widely used as a thermoelectric material¹⁴ and in light-to-heat conversion thin films.¹⁵ Moreover, since bulk β -FeSi₂ is transparent and has a high refractive index of above 5.5 (for wavelength > 1.55 μ m),¹⁶ it is suitable for the application of a thin optical film in the infrared region.¹⁷

To the best of our knowledge, no studies have applied β -FeSi₂ to infrared BPFs. The main reason may be related to the extinction

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coefficient k of the β -FeSi₂. Previously, we developed an excellent solar selective absorption coating using β -FeSi₂ thin films that selectively absorb sunlight using RF magnetron sputtering with a FeSi₂ alloy target.¹⁸ Furthermore, the extinction coefficient k at a wavelength above 2 μm was found to be approximately 0.3, which differs from that of the bulk. To create an effective infrared BPF, it is necessary to eliminate the absorption caused by β -FeSi₂ layers. Previous studies suggest that the difference in k between sputtered films and bulk materials may be attributed to slight variations in composition.¹⁹ Thus, this study aims to produce a β -FeSi₂ thin film with a k value of approximately 0 by controlling the Fe-Si ratio through co-sputtering and developing an infrared BPF with a high and narrow peak at the desired wavelength.

II. MODELING AND DESIGN

A. Modeling for the transmittance and reflectance spectrum of the thin film and the complex reflective index

The optical constants of bulk β -FeSi₂ have been reported in detail by Udono *et al.*,¹⁶ however, there are no reports on the optical constants of polycrystalline β -FeSi₂ thin films. To design the multilayer thin film that meets the BPF requirement, a complex reflective index of the single-layer β -FeSi₂ thin film on Si substrate was obtained as follows. By replacing the film with a single surface shown in Fig. 1, the method of solving the transmittance T_a and reflectance R_a of the air/thin film/substrate part on boundary a using the optical admittance concept is known.²⁰ However, reflection R_b caused by the high refractive index Si substrate at boundary b between the substrate and air also accounts for a large proportion of the measured reflectance. Therefore, the calculated transmittance T and reflectance R spectrum of the thin film were considered using an air/thin film/substrate/air model (Fig. 1).

When calculating n and k of a β -FeSi₂ thin film, light is vertically incident on a single-layer film. On boundary a , reflectance R_a and transmittance T_a as the light passes from air to the substrate and R'_a , T'_a as the light passes from the substrate to air in the opposite direction of the thin film assembly can be computed by the

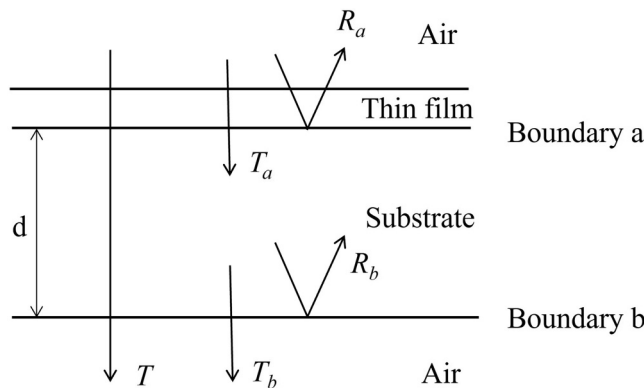


FIG. 1. Optical model of the thin film.

optical admittance concept. Furthermore, R_b and T_b are the reflectance and transmittance, respectively, as light passes from the substrate to the air on boundary b . It is possible to ignore the interference caused by the reflected light at each substrate interface because the thickness of the substrate is significantly greater than the infrared wavelength. Therefore, transmittance T and reflectance R of the entire system can be theoretically calculated using the following equations:

$$R = R_a + T_a T'_a R_b e^{-2\alpha d} + T_a T'_a R'_a R_b^2 e^{-4\alpha d} + \dots$$

$$= R_a + \frac{T_a T'_a R_b e^{-2\alpha d}}{1 - R'_a R_b e^{-2\alpha d}}, \quad (1)$$

$$T = T_a T_b e^{-\alpha d} + T_a T_b R_b R'_a e^{-3\alpha d} + \dots$$

$$= \frac{T_a T_b e^{-\alpha d}}{1 - R'_a R_b e^{-2\alpha d}}, \quad (2)$$

where the substrate's absorption coefficient $\alpha = \frac{4\pi k}{\lambda}$, λ is the wavelength of the incident light, and d is the substrate thickness. n and k values of a single-layer thin film can be obtained by solving the following simultaneous equations using the Newton-Raphson method:

$$\begin{cases} T(n, k) = T_{\text{exp}}, \\ R(n, k) = R_{\text{exp}}, \end{cases} \quad (3)$$

where T_{exp} and R_{exp} are the measured values.

The obtained n and k values are then substituted into R_a and T_a of the multilayer thin film in Eq. (2) to determine the transmittance of the multilayer thin film BPF.

B. Design of the multilayer thin film

We utilized a β -FeSi₂/SiO₂/ β -FeSi₂/quartz three-layer structure to achieve the infrared BPF. The refractive index of the β -FeSi₂ layer was set to 5 based on the calculated results and is explained later in Fig. 3(d). The refractive index of the SiO₂ layer was set to 1.4.²¹ The thickness of each layer was determined by

$$\lambda = 4n_1 d_1 = 2n_2 d_2, \quad (4)$$

where n_1 and d_1 represent the refractive index and thickness of the β -FeSi₂ layer, respectively, and n_2 and d_2 represent those of the SiO₂ layer (see Table I). The transmittance and reflectance of the quartz substrate were determined by Fourier transform infrared spectroscopy (FT-IR) measurements. This analysis is conducted at a desired wavelength of 2 μm , which is in proximity to the bandgap energy of specific PV cells, such as InGaAs. Consequently, photons can be effectively transmitted via the filter at a specific wavelength. The pink curve in Fig. 2(a) shows the transmittance spectrum of the multilayer structure in which the β -FeSi₂ layers have optical constants that are the same as those of the bulk ($k=0$). The calculated transmittance reaches 95%, with a sideband transmittance lower than 5%. The transmittance spectra in other cases, where k is more extensive than zero, are also shown in Fig. 2(a). As the value of k increases to 0.3, there is a significant decrease in the transmittance at the peak. This observation highlights the importance of

TABLE I. Details for each layer of the designed BPFs.

Layer	BPF 1		BPF 2		BPF 3	
	Material	Thickness (nm)	Material	Thickness (nm)	Material	Thickness (nm)
1	β -FeSi ₂	100	Si	151	TiO ₂	208
2	SiO ₂	714	SiO ₂	714	SiO ₂	714
3	β -FeSi ₂	100	Si	151	TiO ₂	208

minimizing the k value of the β -FeSi₂ layer when it is utilized in a BPF. Therefore, reducing the imaginary part of the refractive index is crucial.

Furthermore, we conducted two additional calculations for the BPFs using Si ($n = 3.3$) and TiO₂ ($n = 2.4$) within the same multilayer structure. The details and results are summarized in Table I and Fig. 2(b), respectively. Compared with the β -FeSi₂-based filter, the multilayer film using TiO₂ and Si requires a thicker film with the same number of layers, resulting in a wider full width at half maximum (FWHM) in the transmittance spectrum. Achieving a narrow peak with these materials requires a significant number of layers and an increase in the thickness, leading to higher costs. Therefore, β -FeSi₂ demonstrates better wavelength selectivity and cost reduction due to its higher refractive index.

III. EXPERIMENT

A. Fabrication of the single-layer β -FeSi₂ thin film and its materials characterization

Single-layer β -FeSi₂ thin films were formed on a double-side polished Si substrate ($n \approx 3.3$) using RF magnetron co-sputtering of FeSi₂ and Si targets at high temperatures. The substrate was cleaned by UV/ozone cleaning for 30 min prior to deposition. The

deposition chamber was evacuated to a vacuum of at least 3.0×10^{-4} Pa, and subsequently the substrate was heated. After maintaining the substrate temperature above 430 °C for at least 1 h, pure Argon gas was introduced at a flow rate of 9 sccm and the pressure was maintained at 0.86 Pa. The sputtering process was initiated after cleaning the surfaces of the FeSi₂ and Si targets for approximately 5 min. The applied power of the FeSi₂ target was kept constant at 100 W (sputtering yield in this condition was approximately 8.0 nm/min), and the composition ratio of Fe to Si was controlled by adjusting the applied power of the Si target. The sputtering yield was approximately 0.8 nm/min as the applied power of the Si target was 12 W. The thickness of the β -FeSi₂ thin film was fixed at 100 nm by adjusting the deposition time.

Transmittance and reflectance spectra in the infrared region were measured using FT-IR on Shimadzu IRAffinity-1, and the absorbance spectra were calculated using the following equation: "Absorbance = 1 - Transmittance - Reflectance." The parts of spectra in the wavelength ranging nearby 1.8 and 4.3 μ m were ignored to remove the effects of CO₂ and H₂O in the air. X-ray diffraction (XRD) was based on 2θ scans at an incident angle of 0.5° using Cu K α radiation (Rigaku, Smartlab). X-ray photoelectron spectroscopy (XPS) analysis using Al radiation (ULVAC-Phi, PHI 5000) was used to confirm the chemical state of the elements and

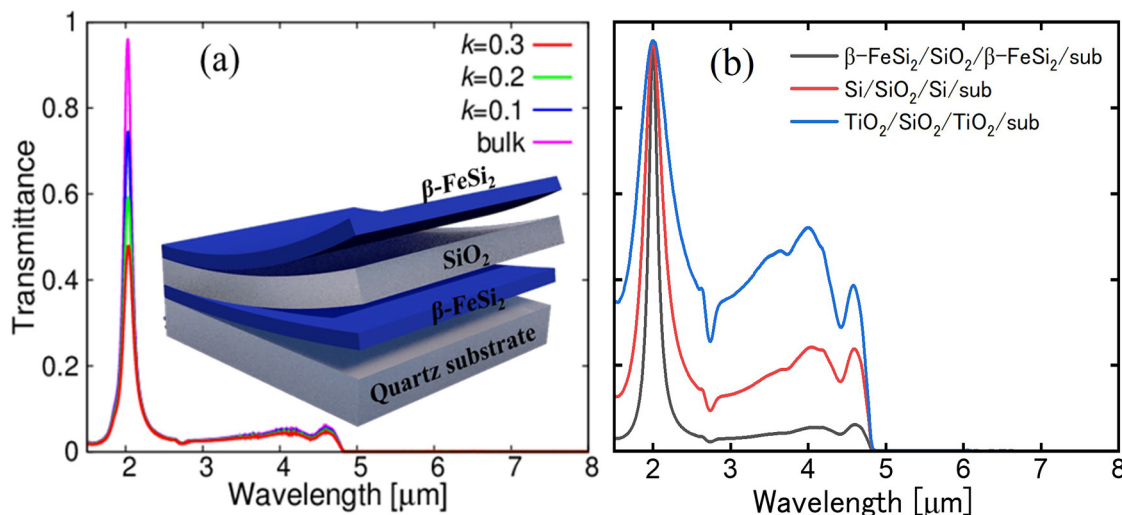


FIG. 2. Transmittance spectrum of the multilayer structure calculated for (a) various values of k representing the β -FeSi₂ layer and (b) BPFs using other high refractive index materials.

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the composition of the thin film. The background types applied to the Fe2p and Si2p regions were Shirley and linear, respectively. The microstructures were examined by scanning electron microscopy (SEM; Hitachi High Tech SU-8020).

B. Fabrication of the multilayer thin film

The β -FeSi₂/SiO₂/ β -FeSi₂ multilayer films were prepared on a 1-mm-thick quartz substrate. The substrate was cleaned by ultrasonic cleaning in acetone for 10 min and by UV/ozone cleaning for 30 min before deposition. The β -FeSi₂ layers were produced in the same manner as described above. The SiO₂ layer was produced after the completion of the first β -FeSi₂ layer. After the heating was stopped and the substrate was returned to room temperature, pure Argon gas was introduced at a flow rate of 9 sccm and the pressure was changed to 1.0 Pa. Sputtering was initiated after cleaning the surface of the SiO₂ target for approximately 10 min. The applied power of the SiO₂ target was fixed at 150 W during the sputtering deposition process. The second β -FeSi₂ layer was repeated after the completion of the SiO₂ layer.

IV. RESULTS AND DISCUSSION

The typical transmittance, reflectance, and absorbance spectra of β -FeSi₂ thin films with varying applied power of Si target (P_{si}) measured using FT-IR are shown in Figs. 3(a) and 3(b). The

refractive index n and extinction coefficient k determined from the transmittance and reflectance, respectively, are shown in Figs. 3(c) and 3(d). As shown in Figs. 3(a) and 3(c), when $P_{si}=0$ W, the β -FeSi₂ thin film exhibited a high absorbance in the infrared region. Its refractive index n and extinction coefficient k values are approximately 5.0 and 0.3, respectively, similar to our previous research.¹⁸ In Fig. 3(b), when $P_{si}=12$ W, the absorbance of the β -FeSi₂ thin film decreased significantly and was almost 0 above the wavelength of 2 μ m. Figure 3(d) shows that the value of k was less than 0.1. To determine how the absorbance of β -FeSi₂ thin film varied with P_{si} from 0 to 20 W, we increased P_{si} by 2 W, measured the optical properties of these samples, and performed replication experiments at $P_{si}=12$ W to provide support for the reliability of the results. By adjusting the sputtering time, the sample thickness was maintained at approximately 100 nm. The absorbance of the samples at a wavelength of 2 μ m is summarized in Fig. 3(e). For $10 \text{ W} \leq P_{si} \leq 12 \text{ W}$, a β -FeSi₂ thin film with an absorbance of about 0 can be produced successfully. The β -FeSi₂ thin film absorbs the infrared wavelength light because of the smaller content of Si in the polycrystalline β -FeSi₂ thin film is smaller than an amount of stoichiometry (Fe: Si = 1:2) at $P_{si} < 12$ W. The addition of an appropriate amount of Si forces stoichiometry to balance and improves the quality of the thin film. When the Si content was excessive, as P_{si} was higher than 12 W, the absorption of the film began to increase.

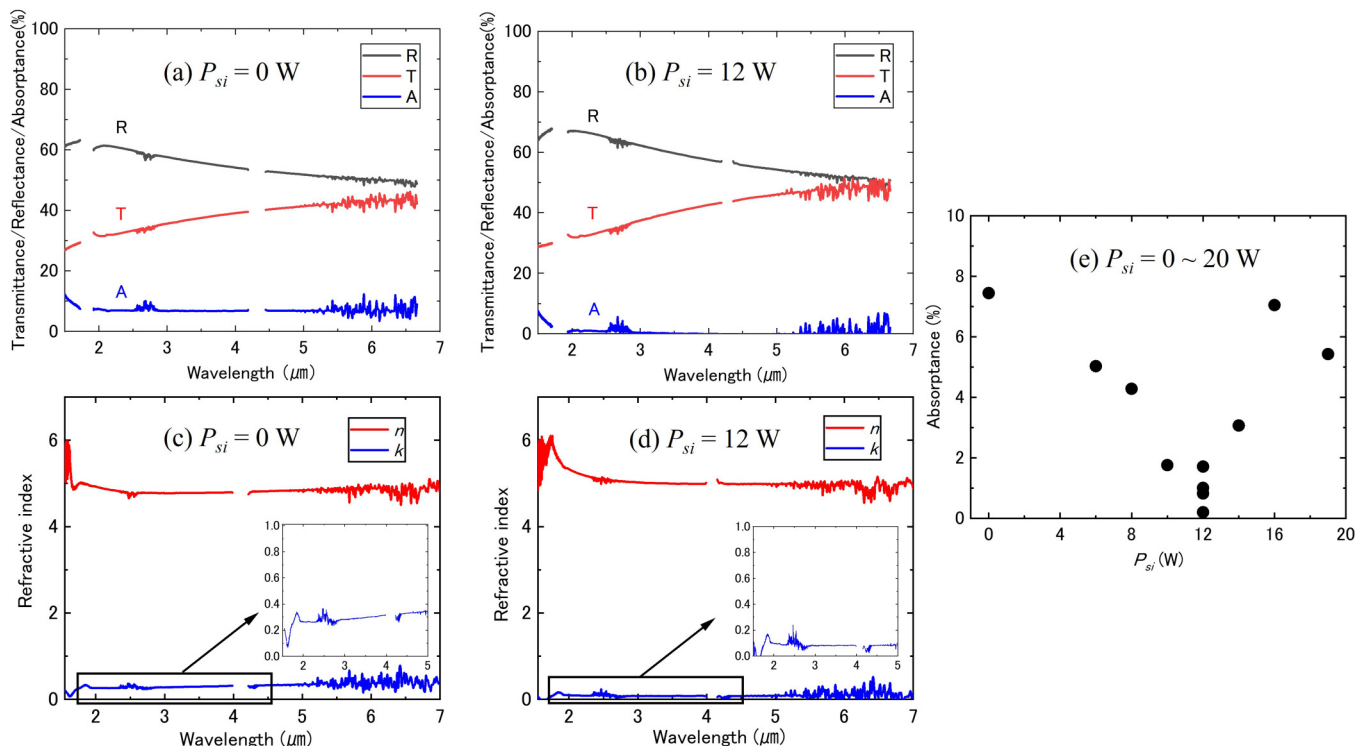


FIG. 3. Optical properties of β -FeSi₂ thin film with varying P_{si} . Transmittance (T), reflectance (R), and absorbance (A) spectra of β -FeSi₂ thin film for (a) $P_{si} = 0$ W and (b) $P_{si} = 12$ W. n and k for $P_{si} =$ (c) 0 W and (d) 12 W. (e) Absorbance at wavelength 2 μ m of the β -FeSi₂ thin film prepared at different P_{si} .

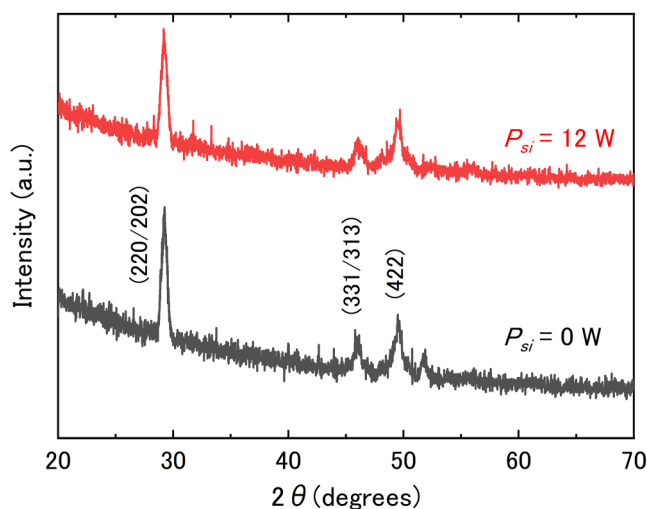


FIG. 4. XRD patterns of β -FeSi₂ thin film prepared when $P_{\text{Si}} = 0$ and 12 W.

XRD and XPS analyses were performed to better understand how the structure and composition of the β -FeSi₂ thin film changed with the Si content. The XRD analysis results are shown in Fig. 4. In each sample, peaks belonging to the orthorhombic²² β -FeSi₂ can be successfully observed, whereas no significant difference in peak position and height was observed. Thus, it can be concluded that the compositional changes in this experiment were small and did not significantly affect the crystallinity.

The concentrations of Si atoms from the XPS quantitative analysis of the β -FeSi₂ thin film prepared at $P_{\text{Si}} = 0$ and, 12 W were 64.1% and 67.2%, respectively. The results showed that the Si concentration measured in all samples was less than the calculated

value when all the atoms deposited on the substrate were incorporated into the thin film. Moreover, the Si concentration was approximately 1:2 when $P_{\text{Si}} = 12$ W. This implies that the composition of Si in the β -FeSi₂ thin film prepared with the FeSi₂ alloy target was smaller than the stoichiometric value; however, the Si atoms supplied from the Si target compensated for the loss of Si atoms caused by reactive evaporation. Hence, for $P_{\text{Si}} = 12$ W, the amount of Si added is precisely equal to the loss amount, and the ratio of Fe: Si in the β -FeSi₂ thin film returns to 1:2, which is the same as the actual stoichiometric value. This also explains why the lowest absorption was achieved in this situation. Therefore, we decided to use a β -FeSi₂ thin film with suitable stoichiometry prepared during the deposition, as mentioned above, for subsequent multilayer film research.

Figure 5(a) shows the transmittance spectra of the multilayer thin films fabricated under $P_{\text{Si}} = 0$ and 12 W, as measured using FT-IR. Compared with the calculation results in Fig. 2(a), the measured spectra are almost identical to that calculated for $k = 0.3$. At $P_{\text{Si}} = 12$ W, owing to the addition of Si atoms, the absorbance of the β -FeSi₂ layer was reduced, and a BPF with a transmittance at the peak exceeding 80% was achieved. Finally, Fig. 5(b) shows that a multilayer structure was properly formed. In other studies, Szeghalmi *et al.*²³ utilized a 15-layer TiO₂/Al₂O₃ structure to attain a peak transmittance of 70% within a BPF. Similarly, Ajith Kumar *et al.*²⁴ and Sullivan *et al.*⁹ achieved satisfactory BPF performance using 43-layer and 10-layer Si/SiO₂ structures, respectively. Pérez *et al.*²⁵ employed ten layers of Ge/SiO to achieve a BPF with a peak transmittance of 70%. Comparatively, the BPF we developed exhibited a comparable peak transmittance while featuring a simpler structure with only three layers and a thinner film thickness.

The position of the BPF peak in this multilayer film structure can be easily controlled by adjusting the thickness of each layer. To demonstrate this, we tested the BPFs at other wavelengths using Eq. (4). The transmittance spectrum of only the peak part of the

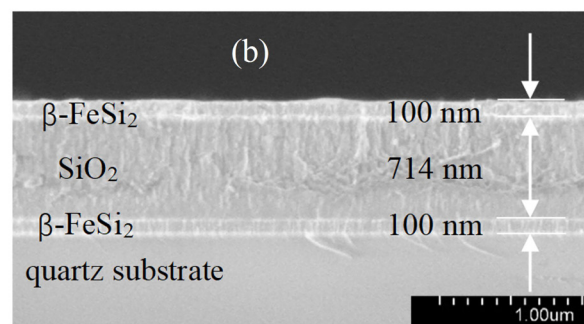
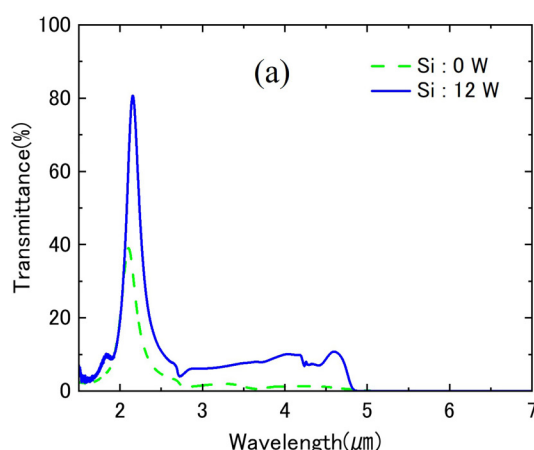


FIG. 5. (a) Transmittance spectrum of the multilayer thin films fabricated under the condition of $P_{\text{Si}} = 0$ and 12 W. (b) Cross-sectional view SEM image of the multilayer thin film.

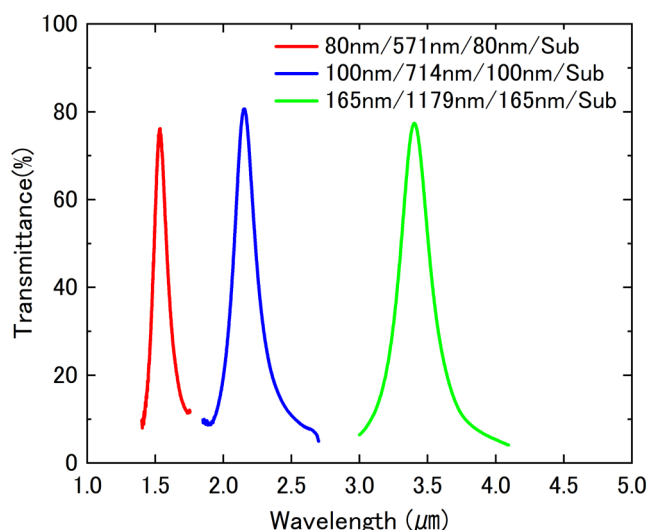


FIG. 6. Transmittance spectrum of the peak part of the BPFs at 1.6, 2, and 3.3 μm wavelength.

BPFs at 1.6, 2, and 3.3 μm wavelength is shown in Fig. 6. The transmission peaks of the produced BPFs all exceeded 75%. Therefore, it can be concluded that this multilayer structure can produce BPFs that perform well in the wavelength range from 1.55 μm ($\beta\text{-FeSi}_2$ bandgap) to 3.5 μm (point where the transmittance of the quartz substrate begins to decrease).

Our results show that $\beta\text{-FeSi}_2$ is a suitable candidate material for infrared BPFs. Owing to the high n , and the low k achieved by adding Si atoms, the BPFs produced using this multilayer film structure achieved the expected narrow and high peaks with a small number of layers and a significantly thin film. Further improvements in the peak transmittance can be achieved by selecting other intermediate layer materials and exploring more suitable film-forming conditions. In addition, because of the high heat resistance and oxidation resistance of $\beta\text{-FeSi}_2$, the infrared BPF prepared using this material is expected to be practically applicable in TPV, infrared heaters, and other fields.

V. CONCLUSIONS

This study investigated the optical properties of $\beta\text{-FeSi}_2$ using co-sputtering, and the performance of BPFs fabricated using this material. Our results demonstrate that, under specific film-forming conditions, the most suitable condition for achieving a transparent $\beta\text{-FeSi}_2$ layer in the infrared region is when the applied power of the Si target is 12 W. Furthermore, the BPFs prepared with the $\beta\text{-FeSi}_2$ layer under this condition exhibited the highest transmittance, exceeding 80%. The experimental results demonstrate that $\beta\text{-FeSi}_2$ is a promising material for the fabrication of infrared BPFs. Using such filters is expected to significantly enhance the efficiency and reduce losses in a variety of applications, such as TPV and infrared heaters.

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AUTHOR DECLARATIONS

Conflict of interest

The authors have no conflicts to disclose.

Author Contributions

Xuanwei Zhang: Investigation (equal); Methodology (equal); Writing – original draft (equal). **Kyoko Namura:** Conceptualization (equal); Investigation (equal); Methodology (equal). **Motofumi Suzuki:** Conceptualization (equal); Investigation (equal); Supervision (equal); Validation (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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