Broadband and perfect absorption in monolayer MoS$_2$-based multilayered structure arranged in Kolakoski sequence

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Abstract
Monolayer MoS$_2$ has excellent optical properties, but its low optical absorption hinders its application in optical absorbers. To improve its absorption performance, we design an aperiodic multilayer structure with monolayer MoS$_2$, the dielectric materials Si and SiO$_2$ arranged by Kolakoski sequence, and achieve the perfect absorption in the visible wavelength range because of its omnidirectional reflectivity. By optimizing the thicknesses of SiO$_2$ ($d_b$), Si ($d_c$) and the repetition number ($N$) of the unit cell, the absorption of the aperiodic multilayer structure can be greatly improved at normal incidence. When $N$ is 3, this aperiodic multilayer structure achieves high absorption of up to 98% in the wavelength range from 350 to 450 nm. As $N$ increases to 5 or 7, the average absorption of this aperiodic multilayer structure exceeds 90% in the 350–600 nm wavelength range. In addition, we verify that the broadband and high absorption obtained in this aperiodic multilayer structure is robust against the angle of incidence and polarization of the incident wave. Our results suggest an alternative configuration with monolayer MoS$_2$ to realize the broadband and perfect absorption, which is useful for future applications in solar cell or photodetectors.

1 Introduction
In recent years, monolayer transition metal dichalcogenides (TMDC$_3$), as two-dimensional (2D) layered materials, have become the hot candidates in the applications of the optoelectronic fields due to their unique electronic and optical properties, such as strong spin–orbit coupling and the apparent excitonic effect [1–11]. Compared with other 2D layered materials, monolayer molybdenum disulfide (MoS$_2$) has unique valley-dependent properties due to the lack of spatial inversion symmetry, such as valley-polarized photoluminescence, valley Zeeman effect, and valley Hall effect [12–15]. In addition, monolayer MoS$_2$ has a hexagonal crystal structure similar to monolayer graphene, while it is a direct bandgap semiconductor and its forbidden bandwidth is 1.8 eV [16]. Based on the above characteristics, monolayer MoS$_2$ plays a significant role in the fields of electronic and optical devices, such as transistors, supercapacitors, gas sensors, biomedical devices, solar cell, and photodetectors [3–7, 15].

It has been reported that monolayer MoS$_2$ has three absorption peaks in the visible wavelength range, whose magnitudes are 23%, 6%, and 7%, respectively [17]. Although the absorption of a free-standing monolayer MoS$_2$ can be 23%, it still cannot meet the needs of optical devices. Many theoretical and experimental researchers have made considerable efforts to solve this problem. For example, Nie proposed an all-dielectric subwavelength grating structure with monolayer MoS$_2$, which enables the perfect light absorption at the wavelength of 586.6 nm with ultra-narrow bandwidth [18]. Zheng designed a metamaterial absorber using monolayer MoS$_2$, V$_2$O$_5$, and a dielectric substrate. They found that this structure has six absorption peaks and their magnitudes can reach more than 90% in the visible wavelength range under the transverse electric (TE) wave [19].

Greatly enhanced absorption with the narrow bandwidths has been achieved in the above structures with monolayer MoS$_2$. However, the broadband and high absorption are more desirable for realistic applications. Researchers have proposed to
combine monolayer MoS$_2$ with various structures to improve their optical absorption, such as photonic crystal structures, metallic microstructures and grating structures. For example, Sun proposed a universal configuration consisting of monolayer MoS$_2$ and the metal–insulator–metal structure, whose average light absorption can reach 74% in the visible spectrum [20]. Huang et al. designed a dielectric grating structure composed of GaN, metallic silver, sapphire, and monolayer MoS$_2$, which can achieve an average absorption rate of 70% [21]. What’s more, Song et al. proposed a multilayer structure composed of Ag rod/monolayer MoS$_2$/dielectric/Ag, which can achieve the broadband and high absorption (more than 80%) in the visible wavelength range [22]. Luo et al. used another multilayer absorber structure consisting of metamaterial, gold, and monolayer MoS$_2$ to achieve the broad and decent absorption, whose magnitudes are over 94% within the wavelength of 594–809 nm [23]. Chen et al. designed a broadband near-infrared absorber composed of Al, SiO$_2$, VO$_2$, and monolayer MoS$_2$ and its average absorption can reach 96.6% within the wavelength range of 800–1160 nm at 25 °C [24]. These studies suggest that monolayer MoS$_2$-based multilayer and grating structures may enable the broadband and significant absorption.

In recent years, there are many investigations on the optical properties in the quasi-periodic structures. Many scientists have designed the various quasi-periodic systems and confirmed that they show the superior optical properties because of their multiple scattering and interference phenomena [25, 26]. For example, Agarwal et al. studied the transmitted properties of the quasi-periodic Fibonacci structure and found that it can be used to manufacture high-sensitivity filters [26]. Compared to periodic and quasi-periodic structures, the optical properties of aperiodic structures have been seldom studied due to their complexity. The Kolakoski sequence is one of the aperiodic sequences, and the previous study has shown that the multilayer structure arranged by this sequence has special optical properties, such as a completely transparent state and the obvious omnidirectional reflectivity [27]. We are wondering whether the aperiodic structure arranged by the Kolakoski sequence can be used to improve the absorption of monolayer MoS$_2$.

On the other hand, the dielectric materials of SiO$_2$/Si are always used in the nanostructures with monolayer MoS$_2$ to improve their absorption performance. Ansari et al. found that TMDC and SiO$_2$/Si heterostructure can achieve more than 30% absorption within a broadband wavelength range and a wide incidence angle window [28]. In addition, Kim et al. revealed that the absorption of air/monolayer MoS$_2$/SiO$_2$/Si substrate can be effectively changed by adjusting the thickness of SiO$_2$ [29]. Considering that they are the most popular components in monolayer MoS$_2$-based absorbers, we also adopt SiO$_2$ and Si as the components to design the broadband optical absorbers.

In this work, we design an aperiodic multilayer structure, whose basic unit cell is composed of monolayer MoS$_2$, Si, and SiO$_2$ arranged by the Kolakoski sequence (it is referred to as the K structure for convenience). We investigated its optical absorption properties using the transfer matrix method and found that by optimizing the number of unit cell repetitions and the thickness of Si and SiO$_2$, the K structure can achieve broadband and high absorption at normal incidence in the visible wavelength range. The broadband and high absorption obtained in this structure can be explained by the critical coupling theory. In addition, we also confirm that this K structure still maintains the broadband and high absorption characteristics at other incident angles for both TE and transverse magnetic (TM) waves. Our work indicates that the K structure presented here can largely enhance the absorption of monolayer MoS$_2$, and that it supports both broadband and high absorption, which overcomes the low absorption of free-standing monolayer MoS$_2$ and contributes to the development of various high-performance optical devices.

2 Basic method and theoretical parameters

The K structure we designed can be simply denoted as air/$S^N$/air, where $S$ is the basic unit cell and it is composed of three materials, including monolayer MoS$_2$ (labeled as A), SiO$_2$ (labeled as B) and Si (labeled as C). $N$ is the number of repetitions of the unit cell. The thicknesses of the materials A, B, and C are characterized by $d_A$, $d_B$, and $d_C$, respectively. Figure 1 is an example of the aperiodic structure and the sequence of materials A, B, and C in the unit cell is determined by the Kolakoski sequence. The Kolakoski sequence

![Fig. 1 A schematic diagram of the K structure, where the black part is monolayer MoS$_2$, the light blue part is SiO$_2$, and the red part is Si.](image-url)
is an infinite sequence composed of two elements, which has obvious “self-describing” characteristics. The specific rule is that the length of the n-th entry of the sequence is equal to the length of the n-th group of the sequence, and the consecutive identical entries in the sequence are defined as a group. Assuming that the Kolakoski sequence consists of \( \{1, 2\} \) and the initial elements are \( a(1) = 1, a(2) = 2 \), the Kolakoski sequence is expressed as

\[
\begin{align*}
1 & 22 11 2 1 22 1 \ldots \\
1 & 2 2 1 1 1 2 2 1 1 2 2 1 1 \ldots 
\end{align*}
\]

If the initial elements are \( a(1) = 2, a(2) = 2 \), the Kolakoski sequence then becomes

\[
\begin{align*}
22 & 11 2 1 22 1 22 \ldots \\
2 & 2 1 1 2 1 2 2 1 2 
\end{align*}
\]

Here, we take the first ten terms of these two sequences and then replace element 1 by \( AB \) and element 2 by \( C \). The sequence then becomes

\[
\begin{align*}
ABCCABABCAB \quad \text{and} \quad CCABABCABCCABCC \\
\text{(it is referred to as the KS structure later), respectively. The TE or TM wave} \\
\text{incidence on the air/SiN/air structure at an angle of} \; \theta. \; \text{The refractive} \\
\text{index of SiO}_2 \; \text{depends on the wavelength of the} \; \lambda, \; \text{and its calculation formula is as follows} \; \text{[30]:}
\]

\[
n_{\text{SiO}_2}(\lambda) = \sqrt{\varepsilon_\beta} = \left(1 + \frac{0.6961663\lambda^2}{\lambda^2 - 0.0684043^2} + \frac{0.4079426\lambda^2}{\lambda^2 - 0.1162414^2} + \frac{0.8974794\lambda^2}{\lambda^2 - 9.896161^2}\right)^{\frac{1}{2}} \]

\[
(1)
\]

In the wavelength range of 390–780 nm, we take the refractive index of Si as 3.42 [12]. The dielectric constant of monolayer MoS\(_2\) at room temperature can be obtained from the following formula [31]:

\[
\varepsilon_A = \varepsilon_{\text{MoS}_2}(\omega) = \varepsilon_{1, \text{MoS}_2} + \sum_{p=1}^{4} \left[ \frac{(f_p + ig_p)\omega_p}{\omega_p + i\gamma_p + \omega} + \frac{(f_p - ig_p)\omega_p}{\omega_p - i\gamma_p - \omega} \right]
\]

\[
(2)
\]

The relevant parameters in this formula can be found in Ref. [31].

In this paper, the transfer matrix method is used to calculate the absorption of the K or KS structure. For the periodic or aperiodic multilayer structure, the total transmission matrix is obtained by combining all the single dielectric

layer characteristic matrix together. For the k-th layer of the multilayer structure, the transmission matrix method can be expressed as

\[
M_k = \left[ \begin{array}{cc} \cos \delta_k & -\frac{i}{2} \sin \delta_k \\ -iq_k \sin \delta_k & \cos \delta_k \end{array} \right] 
\]

\[
(3)
\]

where \( \delta_k = \frac{2\pi}{\lambda_0} n_k h_k \cos \theta_k \), \( n_k, h_k \) and \( \theta_k \) are the refractive index, the thickness and the refractive angle of the k-th layer, respectively. For the different types of the polarized waves, \( q_k \) has different expressions and the details are as follows:

\[
q_k = \begin{cases} 
\sqrt{\mu_0 n_k} \cos \theta_k & \rightarrow \text{TE} \\
\sqrt{\mu_0 n_k} \cos \theta_k & \rightarrow \text{TM}
\end{cases}
\]

\[
(4)
\]

where \( \varepsilon_0 \) and \( \mu_0 \) are the permittivity and permeability of vacuum, respectively. The total transmission matrix is obtained by multiplying the transmission matrix of each layer together, and its expression is as follows:

\[
M = \prod_k M_k = \left[ \begin{array}{cc} m_{11} & m_{12} \\ m_{21} & m_{22} \end{array} \right]
\]

\[
(5)
\]

By combining the boundary conditions, the reflection coefficient \( (r) \) and transmission coefficient \( (t) \) can be calculated by

\[
r = \frac{(m_{21} + m_{22}q_{\text{sub}})q_0 - (m_{21} + m_{22}q_{\text{sub}})q_0}{(m_{21} + m_{22}q_{\text{sub}})q_0 + (m_{21} + m_{22}q_{\text{sub}})}
\]

\[
(6)
\]

\[
t = \frac{2q_0}{(m_{21} + m_{22}q_{\text{sub}})q_0 + (m_{21} + m_{22}q_{\text{sub}})}
\]

\[
(7)
\]

where \( q_0 \) and \( q_{\text{sub}} \) are the corresponding parameters of the cover layer and the substrate layer, respectively. Then, the reflectance \( (R) \), transmittance \( (T) \) and absorptivity \( (A) \) of the structure are, respectively, obtained by

\[
R = |r|^2, \quad T = \left| \frac{q_{\text{sub}}}{q_0} |t| \right|^2 \quad \text{and} \quad A = 1 - R - T
\]

\[
(8)
\]

In general, the absorption of the structure greatly depends on the thicknesses of the components in the unit cell and it is also is influenced by \( N \). Experiment work has shown that the thickness of monolayer MoS\(_2\) \( (d_p) \) is about 0.6 nm [1, 32], while the thicknesses of SiO\(_2\) \( (d_p) \) and Si \( (d_p) \) will be optimized to achieve the good absorption performance of the aperiodic multilayer structure within the operating wavelength (300–700 nm).
3 Analysis and discussion of the results

With the relevant optical parameters of each layer material in mind, we performed the absorption of the K structure as a function of wavelength by the transmission matrix method, where the number of repetitions, $N$, is also a variable parameter for obtaining the broadband and high absorption. Figure 2 shows the variation of optical absorption with the wavelength in the K structure at normal incidence. The different color lines represent the absorption results of the K structure with different $d_b$, $d_c$ and $N$, respectively. The brown line is the absorption of the K structure at $d_b$ = 69 nm, $d_c$ = 30 nm and $N$ = 3 (labeled as model 1), the blue line is the absorption of the K structure at $d_b$ = 70 nm, $d_c$ = 70 nm and $N$ = 5 (labeled as model 2), the yellow line is the absorption of the K structure at $d_b$ = 66 nm, $d_c$ = 75 nm and $N$ = 7, respectively. In particular, the wavelength-dependent optical absorption in air-monolayer MoS$_2$-air structure is also given in this figure (purple line). Compared with the absorption results of air-monolayer MoS$_2$-air and the K structure, we can see that the K structure has achieved the broadband and high absorption characteristics for different $N$. We would like to note that the difference of the absorption in the K structure between $N$ and $N+1$ is not obvious, i.e., the absorption results in the K structure when $N$ = 4 (6) is similar to that when $N$ = 3 (5), and thus the repetition number is increased by two in our calculation. In detail, the absorption spectrum of the K structure shows an overall increasing trend with the increase of $N$. When the wavelength of the incident wave stays within 300–360 nm, the absorption spectrum of the K structure has an oscillating behavior, and the number of the oscillations with the wavelength increases significantly when $N$ is increased. It should be noted that all models 1, 2 and 3 have near perfect absorption, with their average absorption exceeding 95% at wavelengths of 350–450 nm of the incident wave. For example, for model 1 ($N$ = 3), the K structure achieves almost perfect absorption within the wavelength range from 350 to 450 nm, but its absorption becomes poor when the wavelength moves to 450–650 nm. For model 2 ($N$ = 5) or model 3 ($N$ = 7), the K structure shows the slight variation in the absorption spectrum in the wavelength of 350–450 nm, but its average absorption within 350–650 nm increases from 81.59 to 90.44% (91.97%), respectively. It is clear that the increase of the average absorption is more obvious when $N$ increases from 3 to 5, while there is no significant improvement in the spectrum of the absorption when $N$ changes from 5 to 7. For the cases of $N$ > 7, the average absorption of the K structure does not change too much and we do not discuss it here. Furthermore, at wavelengths larger than 672 nm, the optical absorption of the three models almost coincide and show a clear decreasing trend with wavelength, indicating that the variation of $N$ does not significantly affect the absorption features in the long wavelength region.

We can use the theory of critical coupling to explain the cause of the K structure to achieve the broadband and high absorption. A conventional critically coupled resonator is an absorbing structure consisting of an absorbing material and a reflecting cavity, where the light is injected and critically coupled to the reflected light at the incident interface when their phase difference is $\pi$. Then, a standing wave is formed in the absorbing structure and thus the coherent total absorption is achieved [33]. In our structure, the aperiodic multilayer structure with Si and SiO$_2$ constructed by the first kind of the Kolakoski sequence has the omnidirectional reflectivity spectrum in the visible wavelength range and can act as a reflecting cavity. At the same time, the lossy nature of monolayer MoS$_2$ induces absorption in the K structure.

Figure 3 shows the electric field distribution as a function of the wavelength and distance. It can be observed that the strong electric field mainly locates at the beginning of the K structure and the position of 200 nm (both of them are the positions where monolayer MoS$_2$ stay) and their positions almost do not change too much with the wavelength. The strong electromagnetic localization increases the interaction between monolayer MoS$_2$ and the K structure, which effectively produces the significantly enhanced absorption. As the electromagnetic waves propagate along the K structure, the electric field strength gradually decays to zero, suggesting their standing wave characters.
To further shed light on the practical value of the K structure in the application of optical absorbers, we have studied its absorption performance for different incident angles and polarization states, whose results are shown in Fig. 4. From Fig. 4a, it is found that with the incident angle increasing and when it is less than 35°, the wavelength where the broadband and high absorption happen in the K structure gets smaller and the corresponding wavelength range does not change significantly. But, when the incident angle is larger than 60°, the wavelength at which the high absorption occurs in this structure gradually shifts to the short wavelength, and the wavelength range of the high absorption gets decreased for the TE polarized wave, which suggests that the K structure has the low absorption at most wavelengths when the incident angle is high. As shown in Fig. 4b, it is found that the K structure has the high absorption (its average value is larger than 90%) within the wavelength range from 300 to 500 nm for the TM polarized light and its absorption spectrum is less affected by the incident angle when the incident angle is less than 45°. Especially, the absorption is basically close to 1 in the wavelength of 350–450 nm when the incident angle changes from 0° to 30°. When the wavelength is larger than 580 nm, the optical absorption of the K structure is significantly attenuated for almost all the incident angles. From Fig. 4c–f, it can be found that with the increase of N, the K structure has a stable increase in the wavelength range where the broadband and high absorption happen at most incident angles for both TM and TE polarized waves. When the wavelength is in the range of 380–500 nm, the absorption spectrum of each model does not change significantly at the low incident angle and their absorption becomes attenuated with an increased incident angle when the incident angle is larger than 60°. When the wavelength is larger than 500 nm, there is also the wavelength window where the high absorption exists for both TM and TE polarized waves. Compared with the absorption results between the TE and TM polarized waves, it can be found that the absorption spectrum of the K structure has a better performance for the TM polarized wave than that for the TE polarized wave when $N=3$ and their wavelength range associated with the high absorption gets larger with the increase of N. Therefore, we can find that the K structure we designed exhibits the stable broadband and high absorption when the TM or TE polarized wave enters our structure within a relatively wide incident angle range, which suggests that the absorption of the K structure is robust against the incident angle and the polarization state of the incident wave.

Considering the fact that the K and KS structures are different, we wonder how the absorption of the KS structure performs and whether the KS structure supports the broadband and high absorption. Figure 5 shows the optical absorption of the KS structure as a function of wavelength under the normal incidence when $N$ is taken as 3 (labeled as model’ 1), 5 (labeled as model’ 2), and 7 (labeled as model’ 3), respectively. From Fig. 5, it can be observed that the KS structure also has the high absorption characteristics, which appears only in the wavelength of 400–550 nm (440–520 nm) and their values are up to 80% for model’ 1 and model’ 2 (model’ 3). In addition, we note that there are small absorption peaks in the absorption spectrum of the KS structure and the number of the peaks changes with $N$ within the wavelength range where the A and B excitonic bands of monolayer MoS$_2$ stay. To clarify this point, we also performed the calculation of the reflectances of the K and KS structures in the absence of monolayer MoS$_2$ at normal incidence as a function of wavelength when $N=3, 5, 7$ and the results are shown in Fig. 6a and b, respectively.

From Fig. 6a, we can see that when $N=3$, the average reflectivity of the K structure can reach 0.95 in the four bands of 325–355 nm, 375–425 nm, 475–525 nm and 575–700 nm. As $N$ increases, the reflectivity of the K structure in the 475–525 nm band decreases substantially and the multiple peaks appear. On the other hand, there is a significant increase in the width of the photonic band gap in the lower wavelength region and a significant decrease in the width of the photonic band gap in the high wavelength region. As shown in Fig. 6b, the average reflectivity of the KS structure in the visible band can reach above 0.9 when $N=3$. For $N=5$, the KS structure still maintains a high average reflectivity, but oscillates around the wavelengths of 350 nm, 425 nm and 575 nm. For $N=7$, although there are oscillating reflectance peaks within several wavelength regions, the average reflectivity of the KS structure decays
significantly and thus the widths of all the reflectance gaps decrease significantly. The coexistence of low reflectance dips in the KS structure in the absence of monolayer MoS$_2$ within this wavelength range and the A and B absorption peaks of monolayer MoS$_2$ lead to its changed absorption peak numbers with the increased $N$. Obviously, the absorption performance in the K structure is better than that in the KS structure. It can be ascribed to the reflectance characteristics of the K structure in the absence of monolayer MoS$_2$, which has the lower reflectance bands and its wavelength range is relatively wider than that of the KS structure. The combined effects from the reflectance characteristic of the K structure, the excitonic effect of monolayer MoS$_2$ and the layer number of monolayer MoS$_2$ contribute to its broadband and high absorption. Thus, the K structure has the superiority in improving the absorption of monolayer MoS$_2$.

4 Summary

In summary, we use three materials (monolayer MoS$_2$, Si and SiO$_2$) to construct the aperiodic multilayer structure based on the Kolakoski sequence for realizing the broadband and high absorption. By optimizing the thicknesses of Si and SiO$_2$ and the repetition number of the unit cell, we obtain the broadband and high absorption of the K structure within the
visible wavelength range. We also find that the absorption properties of the K structure are robust against the incident angle and polarization state of the incident wave. Our results confirm that the absorption of aperiodic multilayer structure is comparable to that of periodic and quasi-periodic structures, providing an alternative platform for improving the absorption of 2D layered materials and facilitating their advancement in the study of aperiodic structures in optical absorbers.

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Declarations

Conflict of interest The authors declare no competing interests.

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