Monolayer MoS\(_2\) as a new kind of two dimensional (2D) semiconductor has elicited significant attention because of its distinctive electronic and optical properties [1–14]. Monolayer MoS\(_2\) exhibits a direct band gap in the visible frequency range [1–4], which is more favorable for optoelectronic applications than graphene in numerous cases. Monolayer MoS\(_2\) has show numerous potential applications in flexible phototransistors, photodetectors, photovoltaics, and signal amplification [5–13]. Notably, the photoresponsivity of monolayer MoS\(_2\) photodetectors can reach 880 A/W, which is 10\(^6\) better than that of the first graphene photodetectors (<0.5 mA/W) [5].

The optical absorbance in monolayer MoS\(_2\) is minimal (< 11\%) due to its ultrathin thickness, which is not conducive to fabrication of photodetectors, solar cells, and optical amplification. Thus, to promote the applications of monolayer MoS\(_2\), the optical absorbance in monolayer MoS\(_2\) waves should be enhanced. In studies of graphene, several mechanisms have been proposed to enhance the absorption of graphene, e.g., periodically patterned graphene, surface plasmon, microcavity, graphene-negative permittivity metamaterials, and attenuated total reflectance, etc [15–23]. The interaction between graphene and optical beams can also be enhanced when the graphene layers are prepared on top of one-dimensional photonic crystal (1DPC) or with resonant metal back reflectors because of the Fabry-Perot cavity effect [24–26]. The proposed structures are very easy to fabricate using existing technology.

In this Letter, the optical absorption of monolayer MoS\(_2\) prepared on top of 1DPC or metal films with a spacer layer and cover layers is investigated theoretically. We find that the absorption of monolayer MoS\(_2\) can be enhanced by nearly four times because of the F-P interference. The absorption of monolayer MoS\(_2\) with 1DPC is slight larger than that of monolayer MoS\(_2\) with metal films. However, the full width at half maximum (FWHM) of the absorption spectrum of monolayer MoS\(_2\) with metal films is much larger than that of monolayer MoS\(_2\) on top of 1DPC. The absorption of monolayer MoS\(_2\) can also be tuned by varying the thickness of spacer layers and cover layers. Our proposal is very easy to implement and has potential important applications in monolayer MoS\(_2\) optoelectronic devices.

The details of the structure are shown in the inset of Fig. 1 (a). The 0.65 nm monolayer MoS\(_2\) is prepared on top of the SiO\(_2\) spacer layer with high-\(\kappa\) dielectric HfO\(_2\) cover layers. A 1DPC or 130 nm silver film is placed at the bottom of the SiO\(_2\) spacer layer as the resonant back reflector (RBR). The 1DPC is composed of alternating MgF\(_2\) and ZnS layers with a total of 8.5 periods. The permittivity for silver film is frequency dependent [27]. The refractive indices of SiO\(_2\), HfO\(_2\), MgF\(_2\), and ZnS at \(\lambda = 550\) nm are \(n_{\text{SiO}_2} = 1.55\), \(n_{\text{HfO}_2} = 1.93\), \(n_{\text{MgF}_2} = 1.38\), and \(n_{\text{ZnS}} = 2.59\), respectively [27].

To model the absorption of monolayer MoS\(_2\) in these structures, the details of the permittivity of monolayer MoS\(_2\) must be obtained first. Numerous methods based on first-principle calculations have been used in the calculations of the optical spectrum of monolayer MoS\(_2\) [28–31]. However, the reported theory results are conflicting. Differences between theoretical results and experiment results have been noted. The permittivity of monolayer MoS\(_2\) can be extracted from experiments with the use of two exciton transition and the band transition [2]. The imaginary permittivity of exciton can be described by Lorentzian function. Neglecting the variation of the transition matrix elements with energy, the band absorption of a 2D semiconductor \(\alpha_b\) can be described by the step function and the corresponding imaginary permittivity is \(\varepsilon_b = n_{\text{ref}} \omega \alpha_b / \omega\), where \(\alpha_b\) is the speed of light in vacuum and \(n_{\text{ref}} = 2.05\) is the transverse component of the static refractive index [30]. Thus, the imaginary part of the total permittivity can be expressed as

\[
\varepsilon_i = \frac{f_{\text{ex}}^A \Gamma_A}{(E_{\omega} - E_{\text{ex}}^A)^2 + \Gamma_A^2} + \frac{f_{\text{ex}}^B \Gamma_B}{(E_{\omega} - E_{\text{ex}}^B)^2 + \Gamma_B^2} + \frac{f_{\text{ex}}^E}{\hbar \omega} \Theta(E_{\omega} - E_g' , \Gamma_{\text{band}}),
\]

(1)

where \(\Gamma_A\) (\(\Gamma_B\)), \(f_{\text{ex}}^A\) (\(f_{\text{ex}}^B\)), and \(E_{\text{ex}}^A\) (\(E_{\text{ex}}^B\)) are the linewidth, equivalent oscillator strength, and transition energy of A (B) excitons, respectively, \(E_g'\) is the
Fig. 1. (a) Schematic of monolayer MoS₂ prepared on top of 1DPC with SiO₂ spacer layer and HfO₂ cover layer. (b) The absorbance of monolayer MoS₂ as a function of the light wavelength without cover layers for different structures: suspended monolayer MoS₂ (black solid line), monolayer MoS₂ with the 1DPC and 106 nm spacer layer (blue dotted line), monolayer MoS₂ with sliver films and 75 nm spacer layer (red dashed line), and monolayer MoS₂ on the on top of thick SiO₂ substrate (green dashed-dotted line).

The band gap of monolayer MoS₂, \( f_b \), is the equivalent oscillator strength of interband transition, \( \Theta(x, \Gamma) = \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{1}{1 + x^2 \omega^2} \, dx \) is the step function with a broadening of \( \Gamma \), where \( \Gamma_{\text{band}} \) is the linewidth of the interband transition. The spin-orbit splitting is negligible in the band absorption because spin-orbit splitting is much smaller than the band gap. The real part of the permittivity of monolayer MoS₂ can be obtained using Kramers-Kronig relations

\[
\varepsilon_r(\omega) = n_r^2 + \frac{1}{\pi} \int_0^\infty \frac{s \varepsilon_i(\omega)}{s^2 - \omega^2} \, ds,
\]

where \( p \) is the principal value integral.

The parameters in Eq. (1) are obtained by fitting the experimental data using the standard transfer matrix method [24]. The results are shown in Table 1 and Fig. 1(b). Although nine fitting parameters are in Eq. (1), these parameters are relatively independent. For instance, the parameters \( \Gamma_A, f_A^B, \) and \( E_A^\omega \) are decided by the absorption peak located at about 1.88 eV. However, the parameters \( \Gamma_B, f_B^A, \) and \( E_B^\omega \) are obtained by fitting the absorption peak located at 2.02 eV. The linewidth of the band transition is much larger than that of the traditional semiconductor quantum well, which may have been caused by the strong coulomb interaction and the strong electron-phonon interaction. The folds of monolayer MoS₂ and the interaction with the substrate may also enhance the linewidth. The fitting \( E_A^\omega \) of the absorption spectra is small, which indicates that it is mainly contributed by the quasiparticle band absorption.

The monolayer MoS₂ has a large imaginary part of permittivity [Fig. 1(c)]. However, the maximum absorbance (i.e., the directly proportional ratio of the radiation absorbed by the monolayer MoS₂ to that incident upon it) of suspended monolayer MoS₂ is only about 9.6% due to its ultrathin thickness. By contrast, when a monolayer MoS₂ is prepared on top of the 1DPC (silver film), the maximum absorbance can be as high as 34.9% (33.5%) [Fig. 1(c)]. Thus, the absorbance of monolayer MoS₂ with 1DPC (silver) film can be enhanced by about 3.64 (3.49) times. In this structures, the monolayer MoS₂ and the RBR act as the mirrors of the F-P cavity. The light propagates back and forth in the F-P cavity, which enhances the absorption of monolayer MoS₂. The maximum absorption of monolayer MoS₂ with 1DPC is slightly larger than that of monolayer MoS₂ with a silver film due to the higher reflectivity of 1DPC. However, the reflectivity of 1DPC is limited to the photonic band gap width in 1DPC. By contrast, the silver film can reflect lights effectively within the visible light range. Thus, the FWHM of the absorption spectrum of monolayer MoS₂ with silver film is much larger than that of monolayer MoS₂ with 1DPC. To contrast, we also show the absorbance of the monolayer MoS₂ prepared on top of thick SiO₂ substrate in fig. 1(d). The maximum absorbance is only about 6.1%, which is smaller than that of suspended monolayer MoS₂ due to that the traditional substrate material enhance the reflection of the monolayer MoS₂.

The absorption of monolayer MoS₂ can also be tuned by varying the thickness of the spacer layer. In Figs. 2(a) and 2(b), we plot the absorbance of monolayer MoS₂ as a function of the wavelength and the spacer layers thickness \( d_s \) without cover layers. Similar to the microcavity, the resonant absorption wavelength of monolayer MoS₂ with 1DPC can be described as \( n_{\text{SiO}_2} \lambda / 4m\pi \) with including the half-wave loss, where \( m \) is an integer. Thus, the absorption peak photon energy increases with decreasing spacer layer thickness. However, the resonant absorption wavelength of monolayer MoS₂ with silver films is not strictly equal to \( n_{\text{SiO}_2} \lambda / 4m\pi \) due to the dispersion and existing skin depth in the silver films. The absorption peak photon energy of monolayer MoS₂ with silver films increases slowly with decreasing spacer layer thickness.

| Table 1. Fitted parameters in Eq. (1) |
|-------------------------------------|
| \( E_A^\omega \) | \( \Gamma_A \) | \( E_B^\omega \) | \( \Gamma_B \) | \( f_A^A \) |
| 1.88-4eV | 28meV | 2.02eV | 42meV | 0.32meV |
| \( f_B^A \) | \( E_g \) | \( \Gamma_{\text{band}} \) | \( f_b \) |
| 0.43meV | 2.43eV | 0.398eV | 59 |
thickness. The electron mobility in monolayer MoS$_2$ can be enhanced several times by the high-$\kappa$ dielectric HfO$_2$ cover layers [32,33]. The cover layers can also adjust the absorption of monolayer MoS$_2$. The details are shown in Figs. 2(c) and 2(d). The normal cover layers cannot enhance the absorptance of monolayer MoS$_2$. However, the peak photon energy can be tuned by varying the thickness of the cover layers. Different from the spacer layer, the absorption of monolayer MoS$_2$ with both 1DPC and silver films are the same way when the cover layer thickness increases.

Finally, we turn to the discussion on the absorptance of the monolayer MoS$_2$ prepared on top of Si substrate with SiO$_2$ spacer layer. This structure has been fabricated in the experiment [5]. The Si substrate can also act as the RBR due to its high refractive index, and thus enhance the absorptance of the monolayer MoS$_2$. The absorptance of the monolayer MoS$_2$ with Si substrate as function of the spacer layers thickness $d_s$ for optical wavelength $\lambda = 561 \text{nm}$ (same as in the experiment [5]). For $d_s = 270 \text{nm}$, the maximum absorptance of the monolayer MoS$_2$ is about 9% [Fig. 3(a)]. This is why a $d_s = 270 \text{nm}$ SiO$_2$ spacer layer is fabricated in experiment. By contrast, the absorptance of suspended monolayer MoS$_2$ is about 5.9% and the absorptance of monolayer MoS$_2$ with silver films is about 22% due to the higher reflectivity of silver films. Furthermore, the FWHM of the absorption spectrum of monolayer MoS$_2$ with metal films is larger than that of monolayer MoS$_2$ with Si substrate [Fig. 3(b)]. Thus if the Si substrate is replaced by the metal films in the experiment, the photoresponsivity and the FWHM of monolayer MoS$_2$ photodetectors can be further enhanced greatly.

In conclusion, the optical absorption of monolayer MoS$_2$ prepared on top of 1DPC or silver films with a spacer layer is investigated theoretically. In these structures, the maximum optical absorptance of monolayer MoS$_2$ can be raised to 35% with a large FWHM due to the F-P cavity effect. The absorptance of monolayer MoS$_2$ with 1DPC is slightly larger than that of monolayer MoS$_2$ with silver films. However, the FWHM of the absorption spectrum of monolayer MoS$_2$ with silver film is much larger than that of monolayer MoS$_2$ with 1DPC. The absorption of monolayer MoS$_2$ can also be tuned by varying the thickness of the spacer layers and cover layers. Our proposal is very easy to implement and may have potentially important applications in the development of monolayer MoS$_2$ based optoelectronic devices.

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References

1. Q. H. Wang, K. Kalantar-Zadeh, A. Kis, J. N. Coleman, and M. S. Strano, Nat. Nanotech. 7, 699 (2012).
2. K. F. Mak, C. Lee, J. Hone, J. Shan, and T. F. Heinz, Phys. Rev. Lett. 105, 136805 (2010).
3. A. Splendiani, L. Sun, Y. Zhang, T. Li, J. Kim, C. Y. Chim, G. Galli, and F. Wang, Nano Lett. 10, 1271 (2010).
4. T. Li and G. Galli, J. Phys. Chem. C 111, 16192 (2007).
5. O. Lopez-Sanchez, D. Lembke, M. Kayci, A. Radenovic, and A. Kis, Nat. Nanotech. 8, 497 (2013).
6. Z. Yin, H. Li, H. Li, L. Jiang, Y. Shi, Y. Sun, G. Lu, Q. Zhang, X. Chen, and H. Zhang, ACS Nano 6, 74 (2011).
7. H. S. Lee, S. W. Min, Y. G. Chang, M. K. Park, T. Nam, H. Kim, J. H. Kim, S. Ryu, and S. Im, Nano Lett. 12, 3695 (2012).
8. W. Choi, M. Y. Cho, A. Konar, J. H. Lee, G. Cha, S. C. Hong, S. Kim, J. Kim, D. Jena, J. Joo, and S. Kim, Adv. Mater. 24, 5832 (2012).
9. M. Bernardi, M. Palummo, and J. C. Grossman, Nano Lett. 13, 3664 (2013).
10. J. Pu, Y. Yomogida, K. K. Liu, L. J. Li, Y. Iwasa, and T. Takenobu, Nano Lett. 12, 4013 (2012).
11. C. Janisch, N. Mehta, D. Ma, A. L. Elas, N. Perea-Lopez, M. Terrones, and Z. Liu, Opt. Lett. 39, 383 (2014).
12. A. Sobhani, A. Lauchner, S. Najmaei, C. Ayala-Orozco, F. Wen, J. Lou, and N. J. Halas, Appl. Phys. Lett. 104, 031112 (2014).
13. X. Gan, Y. Gao, K. F. Mak, X. Yao, R. J. Shiue, A. van der Zande, M. E. Trusheim, F. Hatami, T. F. Heinz, J. Hone, and D. Englund, Appl. Phys. Lett. 103, 181119 (2013).
14. Y. V. Bludov, M. I. Vasilevskiy, and N. M. R. Peres, J. Appl. Phys. 112, 084320 (2012).
15. S. Thongrattanasiri, F. H. L. Koppens, and F. J. G. de Abajo, Phys. Rev. Lett. 108, 047401 (2012).
16. A. Ferreira, N. M. R. Peres, R. M. Ribeiro, and T. Stauber, Phys. Rev. B 85, 115438 (2012).
17. M. A. Vincenti, D. de Ceglia, M. Grande, A. D’Orazio, and M. Scalora, Opt. Lett. 38, 3550 (2013).
18. A. Ferreira and N. M. R. Peres, Phys. Rev. B 86, 205401 (2012).
19. Z. Z. Zhang, K. Chang, and F. M. Peeters, Phys. Rev. B 77, 235411 (2008).
20. W. Zhao, K. Shi, and Z. Lu, Opt. Lett. 38, 4342 (2013).
21. Q. Ye, J. Wang, Z. Liu, Z. C. Deng, X. T. Kong, F. Xing, X. D. Chen, W. Y. Zhou, C. P. Zhang, and J. G. Tian, Appl. Phys. Lett. 102, 021912 (2013).
22. R. Alaee, M. Farhat, C. Rockstuhl, and F. Lederer, Optics Express 20, 28017 (2012).
23. X. Zhu, W. Yan, P. U. Jepsen, O. Hansen, N. A. Mortensen, and S. Xiao, Appl. Phys. Lett. 102, 131101 (2013).
24. J. T. Liu, N. H. Liu, J. Li, X. J. Li, and J. H. Huang, Appl. Phys. Lett. 101, 052104 (2012).
25. N. M. R. Peres and Y. V. Bludov, EPL 101, 58002 (2013).
26. J. T. Liu, N. H. Liu, L. Wang, X. H. Deng, and F. H. Su, EPL 104, 57002 (2013).
27. E. D. Palik, ed., Handbook of Optical Constants of Solids (Academic Press, Boston, 1985).
28. D. Y. Qiu, F. H. da Jornada, and S. G. Louie, Phys. Rev. Lett. 111, 216805 (2013).
29. H. Shi, H. Pan, Y. W. Zhang, and B. I. Yakobson, Phys. Rev. B 87, 155304 (2013).
30. T. Cheiwchanchamnangij and W. R. L. Lambrecht, Phys. Rev. B 85, 205302 (2012).
31. A. Ramasubramaniam, Phys. Rev. B 86, 115409 (2012).
32. B. Radisavljevic, A. Radenovic, J. Brivio, V. Giacometti, and A. Kis, Nat. Nanotech. 6, 147 (2011).
33. Y. Yoon, K. Ganapathi, and S. Salahuddin, Nano Lett. 11, 3768 (2011).