Dual-band absorption enhancement of monolayer transition-metal dichalcogenides in metamaterials*

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Enhancement of light absorption in two-dimensional (2D) single atomic layer materials is a significant issue for applications of 2D material-based optoelectronic devices. In this letter, the dual-band enhanced absorptions in monolayer transition-metal dichalcogenides (TMDs) are obtained based on the metmaterial nanostructures. The proposed nanostructure consists of a monolayer TMDs sandwiched between Ag nanodisks and a dielectric spacer on Ag substrate. The excitations of the surface plasmon polaritons (SPPs) and the magnetic dipole resonances contribute to the high absorption efficiency, and the resulting absorption enhancement can be separately tuned by simply adjusting the structural parameters such as the spacer thickness, the size and the period of the Ag nanodisks. In addition, a hybrid nanostructure consisting of different nanodisks can increase the absorption bandwidth. The calculated results may have some potential applications in photodetection and wavelength-selective photoluminescence.

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Two-dimensional (2D) materials, usually referred to as the single layer materials, such as graphene[1], black phosphorus[2], hexagonal boron nitride[3] and transition-metal dichalcogenides (TMDs)[4,5], manifest unique band structures and outstanding optical properties. They are quite attractive for a wide range of photonics, electronics and optoelectronics applications. Especially for TMDs (MoS_2, $MoSe_2$, WS_2 and WSe_2) have tunable bandgaps ranging from 1 eV to 2 eV. Moreover, TMDs undergo a transition from an indirect to direct-gap semiconductor from bulk to monolayer due to quantum confinement effects[6]. The remarkable direct-bandgap semiconductor characteristic allows the monolayer TMDs to possess important applications in photoluminescence, photodetection and photovoltaic devices[4,7]. Although the optical absorption coefficient is considerably high for a single atomic layer[8], for the optoelectronics and photonics applications, the intrinsic small thickness poses a great challenge for light-material interactions, which therefore results in poor light emission and absorption behavior. Without plasmons excited in monolayers TMDs, the absolute absorption of light in free-standing monolayer TMDs is only about 10%—20%[9] in the visible regime, such as the monolayer MoS_2, it's average single pass optical absorption is about 10%[10]. The intrinsic poor absorption of light in monolayer TMDs limits the effective performance of TMDs-based devices. So it is urgently desired to improve the light absorption in monolayer TMDs and various approaches have been reported. For instance, the one-dimensional photonic crystal or metal films resonant back reflector is used to enhance the absorption of monolayer MoS_2, the maximum absorptance can be raised to 35%[11]. In a multilayers, the absolute absorption up to 96% at the visible wavelengths in atomically thin layer is achieved due to the excitation of Tamm states[12]. The dual band enhanced absorptions of monolayer MoS_2 are also realized by using plasmonic metamaterial[13]. Based on the critical coupling with guided resonances, Li et al realized total absorption in monolayer MoS_2 at λ=679.2 nm both theoretically and numerically[10]. Long et al proposed a magnetic coupling metasurface to achieve broad band and broad angular absorption as high as 72.7% for MoS_2 monolayer within the visible wavelength range[14]. At 619 nm, the absorption of WS_2 monolayer was increased to 61% through plasmonic coupling[15]. Based on the excitations of surface plasmon polaritons (SPPs) and magnetic resonances in metamaterials, the double-band absorption enhancement in monolayer graphene can be realized[16]. In addition, metmaterials provide a significant platform for high-performance photodetection[17] and Photoluminescence[18,19].

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In this letter, the dual-band enhanced absorptions of the monolayer MoS_2, $MoSe_2$, WS_2 and WSe_2 in the metmaterial are investigated, where the monolayer TMDs is sandwiched between a periodic array of Ag nanodisks and a SiO_2 spacer supported on a metal (Ag) substrate. The simulated results show that the enhanced TMDs absorptions originate from the excitation of SPP and magnetic resonance and the absolute absorption of light in TMDs monolayer is improved up to 84.5% at the wavelength of the magnetic resonant peak. The influences of structure parameters such as the spacer thickness, the size and the periodicity of the Ag nanodisks are discussed. In addition, the bandwidth of such high absorption in TMDs monolayer at the magnetic resonance wavelength can be broadened by using hybrid structure with different magnetic resonances.

Fig.1 shows schematically the unit cell of the proposed metamaterial for dual-band light absorption enhancement of monolayer TMDs. The monolayer TMDs are sandwiched between the Ag nanodisks and the SiO_2 spacer (with thickness of d) supported on an Ag substrate. The Ag reflector is assumed to have a thickness of 200 nm. The Ag nanodisk is supposed to lie on the x-y plane. The thickness, period and radius of the Ag array are h, Px, Py and R, respectively. Light is incident in the negative z-axis direction, θ is the angle of incident light. Numerical simulations were performed with a finite element method (COMSOL Multiphysics), the reflection and absorption spectra and the electromagnetic field distributions are calculated. In numerical calculations, the frequency-dependent relative permittivity of Ag is taken from experimental data[20], and the relative permittivity of SiO_2 is set to be 2.1. In simulations, the monolayer TMDs such as MoS_2 whose wavelength-dependent complex permittivity has been measured experimentally by Liu et al[8], it is employed as a thin film with thickness of 0.615 nm. Since the thickness of the silver substrate is much larger than the penetration depth of electromagnetic waves, the transmission from the proposed structure is very close to zero. Then, the absorption of the proposed TMDs-based absorber can be described by $A=1-R$, where R is reflection from the metamaterial structure. To quantitively evaluate the amount of energy that is absorbed by the monolayer transition-metal dichalcogenides and Ag in the composed systems, we performed integration for the power density in the respective materials. Then the absorption in the monolayer transition-metal dichalcogenides can be calculated by further dividing it by the total incoming power.

To clearly illustrate the physical mechanism, we firstly investigate the absorption of the proposed MoS_2-based absorber and the monolayer MoS_2 in the system under illumination of TE-polarized (its polarization along the y-axis direction) normal incident light, as shown in Fig.2. The absorption of monolayer MoS_2 (Ag) represent the power ratio of electromagnetic energy absorbed by monolayer MoS_2 (Ag) in the system and the incident electromagnetic energy. The Ag plasmonic nanodisk arrays possess the effect of localized plasmon resonances[21], since the electric field of TE-polarized light is perpendicular to xz plane, the plasmonic resonance is dependent on Py in this condition. Meanwhile, the magnetic resonances also exist, which results from the resonance excitation of individual Ag nanodisks stacked above the thick silver film with a dielectric spacer. The induced localized plasmonic resonance and magnetic resonance restrain the reflection from MoS_2, and dual-band strong absorption peaks can be realized at the selected resonance frequency. Fig.2 shows the calculated absorption spectra of monolayer MoS_2, Ag and the total metamaterial under normal incidence, where R=65 nm, h=50 nm, d=30 nm, $Px=Py$=500 nm. Two resonance modes are found, which are centered at wavelengths of λ_1=550 nm and λ_2=804 nm. For resonance mode λ_1, the normalized absorptions of monolayer MoS_2, Ag and the total metamaterials are 0.72, 0.2 and 0.92, respectively. For the broad resonance mode λ_2, the corresponding normalized absorptions are 0.995, 0.25 and 0.745, respectively. The absorbed light is mainly dissipated in MoS_2 rather than in Ag. It is well-known that the average absorption efficiency of a suspended monolayer MoS_2 in the optical wavelength rang is about 10%, which limits its optoelectronic applications[22,23]. Obviously, the absorption efficiency of the monolayer MoS_2 in the proposed metamaterial is enhanced noticeably, it has an absorption enhancement of more than 7.5 times. The enhanced absorption of light in monolayer MoS_2 could find some potential applications in optoelectronic devices, such as photodetectors and ultra-thin light source.

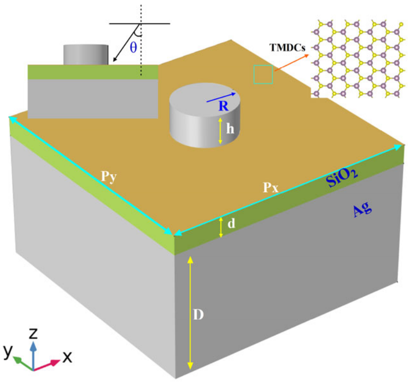

Fig.1 Schematic of the proposed metamaterials for double-band light absorption enhancement of monolayer TMDs

To reveal the physical origin of the absorption in the metamaterial nanostructure, the electromagnetic field distributions for the resonant modes are investigated. Fig.3 illustrates the electric field and magnetic field distributions on the xoy plane across the center of the SiO_2 spacer, in the metamaterial unit $Px=Py$=500 nm, R=65 nm, h=50 nm, d=30 nm at λ_1 and λ_2 labeled in Fig.2. At the resonance wavelength of λ_1, parallel electromagnetic field bands stretching along the y-axis direction are formed, although they are disturbed near the Ag nanodisks, as shown in Fig.3(a) and (b). In fact, such electromagnetic field distributions mainly correspond to the excitation of SPPs[24]. At the resonance wavelength of λ_2, the electric fields are highly confined near the edge of the Ag nanodisks and have two field "hot spots" on the upper and lower sides extending into the SiO_2 spacer. The magnetic fields are concentrated within the SiO_2 spacer and have a maximum under the Ag nanodisks, as shown in Fig.3(c) and (d). Such distribution properties of electromagnetic fields are mainly the typical characteristic of a magnetic resonance[25].

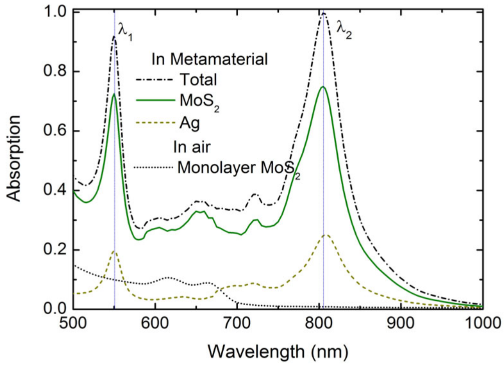

Fig.2 The absorption spectra of the total metamaterials, MoS_2 and Ag in the wavelength range from 500 nm to 1 000 nm under normal incidence, where R=65 nm, h=50 nm, d=30 nm and Px=Py=500 nm

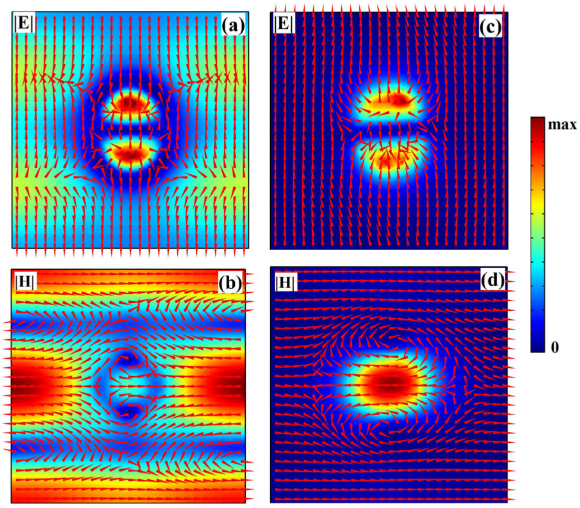

Fig.3 (a,c) The normalized electric field intensity (|E|) and (b,d) the magnetic field intensity (|H|) on the xoy plane across the center of the SiO_2 spacer at λ_1=550 nm and λ_2=804 nm, where red arrows represent the field direction, colors show the field strength, and the electric field is along y-direction

It should be pointed out that the proposed scheme of metamaterials can also be applied to obtain enhanced absorption in other monolayer TMDs, such as $MoSe_2$, WS_2 and WSe_2, not just MoS_2. The simulated absorption spectra for the composed systems with respectively monolayer $MoSe_2$, WS_2, and WSe_2, are demonstrated in Fig.4. The thicknesses of the monolayer $MoSe_2$, WS_2, and WSe_2 are 0.646 nm, 0.618 nm, and 0.649 nm, respectively. The complex permittivity of monolayer TMDs measured experimentally by Liu et al[8] is used for our numerical simulations. Compared with the low absorption spectra of monolayer TMDs suspended in air (the short dotted lines), it is found that the appearance of double absorption peaks within wavelengths ranging from 500 nm to 1 000 nm. The enhanced absorptions based on the excitation of SPP and magnetic resonance occur at the wavelengths of 550 nm (773.7 nm), 550 nm (792.3 nm) and 550.8 nm (805.3 nm), corresponding to the monolayer $MoSe_2$, WS_2 and WSe_2, respectively. The absorption can be 0.696 (0.845), 0.65 (0.774) and 0.70 (0.783).

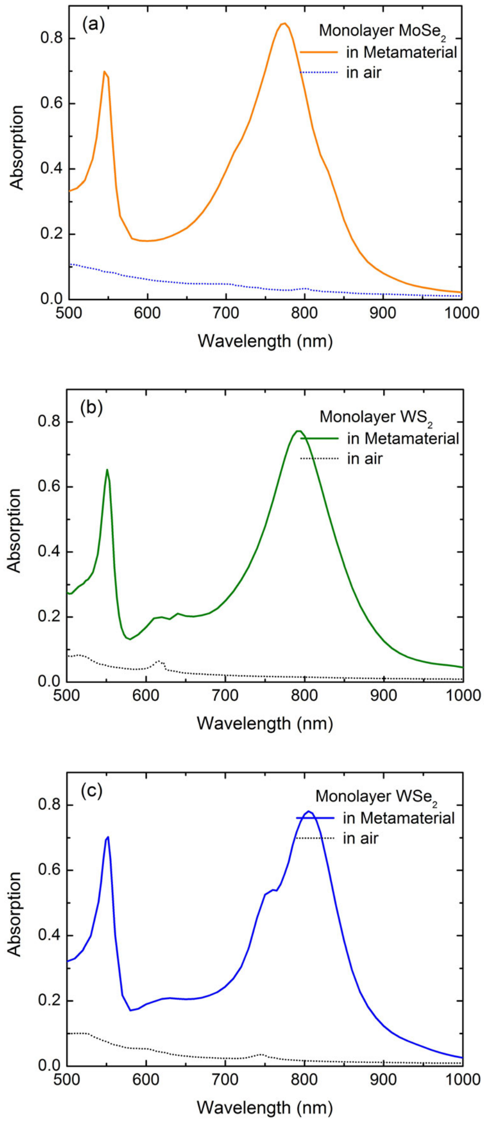

Fig.4 Absorption spectra of monolayer TMDs, such as (a) $MoSe_2$, (b) WS_2 and (c) WSe_2, suspended in air and in metamaterial structures, where the thicknesses of the monolayer $MoSe_2$, WS_2 and WSe_2 are 0.646 nm, 0.618 nm and 0.649 nm, respectively

Next, the influences of geometrical parameters on the absorption are investigated. Fig.5 shows the calculated monolayer MoS_2 absorptions as a function of wavelength and radius R of the nanodisk, the SiO_2 spacer thickness and the periodic under normal incidence, respectively. It is found that the resonance mode λ_1 is almost independent of R, while it increases with the periodic P_y. This is because the resonant wavelength of surface plasmon mode is not sensitive to the size of nanodisks, but mainly depends on the period of the Ag nanodisk array and $P_y \approx \lambda\sqrt{(\varepsilon_{\mathrm{d}}+\varepsilon_{\mathrm{Ag}})/(\varepsilon_{\mathrm{d}}\cdot\varepsilon_{\mathrm{Ag}})}$ [26], ε_{d} and $\varepsilon_{\mathrm{Ag}}$ denote the dielectric constants of the spacer dielectric and metal silver, respectively. At the same time, the monolayer MoS_2 retains the light absorptions of about 70% for the resonance modes λ_1 for the structures with different R. For the second absorption peak λ_2, it is related to a magnetic dipole resonance, whose position determined by both the geometrical and material parameters of the nanodisk and the distance between the Ag nanodisk and Ag film. Fig.5(a) and Fig.5(b) show the effect of the diameter of the Ag nanodisk and the SiO_2 spacer thickness on absorption peaks. It is clear that when the spacer thicknesses (the radius of the nanodisk) are decreased (increased), the resonance mode λ_2 will be red-shifted. This phenomena can be explained by formula[27]: $\lambda_{\mathrm{MP}}=2\pi c[(L_m+L_e)C]^{1/2}$, where c is the speed of light in free space, λ_{MP} is magnetic dipole resonance wavelength, C is the capacitance, which is introduced by the Ag nanodisk and Ag substrate, the capacitance is determined by SiO_2 spacer thickness, contact area between nanodisk and substrate. L_m and L_e are mutual inductance and self-inductance, which are also introduced by the Ag nanodisk and Ag substrate. When the spacer thickness and the radius R reduced or increased, the capacitance C will reduce or increase, for the contact area between nanodisk and substrate is smaller or larger. Therefore, as the spacer thickness increases, the second MoS_2 absorption peaks will be red-shifted, as shown in Fig.5(b). However, the resonance mode λ_2 almost has no shift with the period of sliver nanodisks, since the magnetic resonance wavelength is insensitive to the period of nanodisks as shown in Fig.5(c).

In addition, since all the above results are based on normal incident light, however, in the applications of TMDs-based photonic devices, the proposed metamaterial nanostructures should have high absorption efficiency which is insensitive to the incident angles. For the reason, we illustrate the optical absorption of MoS_2 as a function of incident wavelength and incident angles, as shown in Fig.6. The SPP effect-induced resonance absorption is very sensitive to the incident angle due to its inherent mechanism. It can be observed that the second absorption peak is insensitive to the incident angle, when the incident angle increases to 50°, the absorption peaks of MoS_2 at λ_2 are still higher than 70%. While when the incident angle continues to increase, the absorption intensity becomes weaker and then disappears. The good absorption stability under the relatively wide range of oblique incidence undoubtedly enables this nanostructure to be more feasible in practical applications.

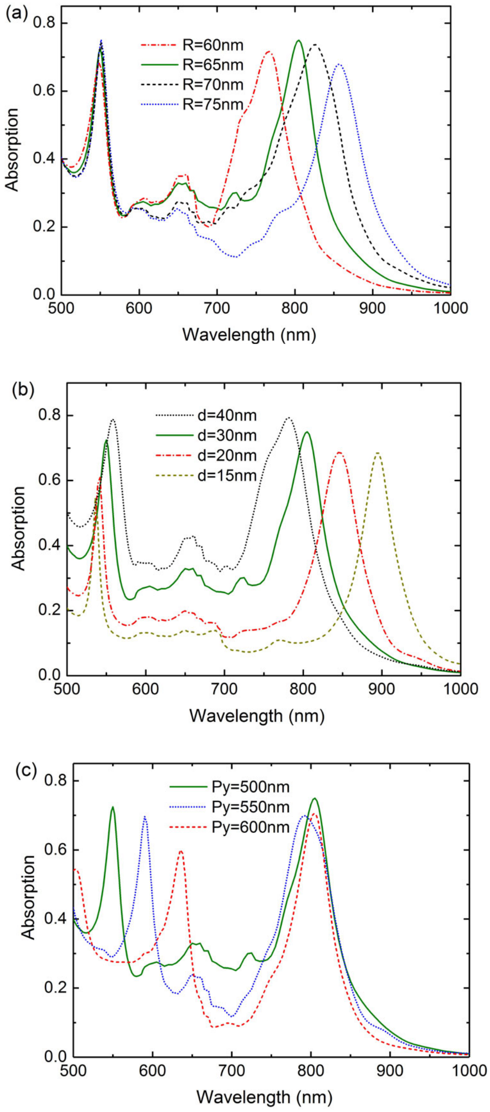

Fig.5 The absorption spectra of monolayer MoS_2 under (a) different *R*, (b) different *d* and(c) different periods *Py* in the wavelength range from 500 nm to 1 000 nm, where the other parameters are the same as those in Fig.2

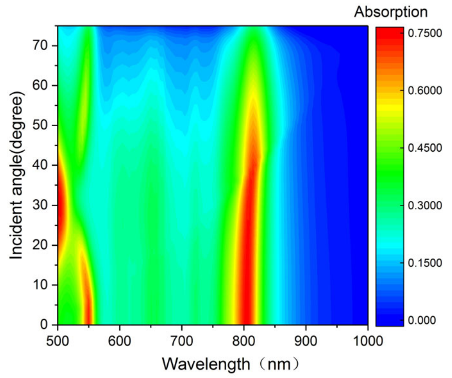

Fig.6 Simulated absorption spectra of monolayer MoS_2 as a function of wavelength and incidence angles for TE

polarization, where the monolayer MoS_2 exhibits stable absorbance and bandwidth over a wide range of oblique incidence at the second resonant mode

It can be clearly seen from Fig.2 that for such single-radius nanodisk, the absorption bandwidth is narrow, even for the magnetic resonance mode, the bandwidth only about 60 nm (absorption over 40%). However, in some practical applications, it is desirable to broaden the absorption bandwidth of TMDs. If we put two different radius nanodisks in one period, forming a double-radius array as shown in the inset of Fig.7, the MoS_2 absorption band width with absorption efficiency over 40% can reach 150 nm. Fig.7 shows the simulated normal incidence absorption spectra in TE polarization as a function of wavelength, where R_1=75 nm, R_2=65 nm (dotted line), R_1=70 nm, R_2=60 nm (solid line), and the other parameters are the same as those in Fig.2. The magnetic field distributions of the hybrid nanostructure at resonance wavelength of λ_A and λ_B are shown in the inset of Fig.7, which are related to magnetic dipole resonances. At resonance wavelength λ_A=803 nm, the magnetic fields are mainly confined to the SiO_2 spacer under the second Ag nanodisk with a radius of R_2, whereas at the resonance wavelength λ_B=862 nm, the magnetic fields are not only highly limited to SiO_2 spacer under the first Ag nanodisk with a radius of R_1, but also some magnetic fields are confined to the SiO_2 spacer under the second one. Briefly speaking, the wide absorption bandwidth of MoS_2 is caused by the overlap of the two magnetic resonances.

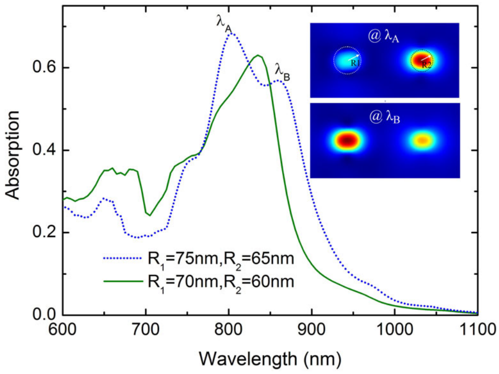

Fig.7 The simulated normal incidence absorption spectra in TE polarization as a function of wavelength for R_1=75 nm and R_2=65nm (dotted line), R_1=70 nm and R_2=60 nm (solid line), where the insets are the magnetic field distributions at λ_A and λ_B respectively

In summary, a hybrid nanostructure consisting of a monolayer TMDs (MoS_2, $MoSe_2$, WS_2 and WSe_2) sandwiched between Ag nanodisk metamaterials with SiO_2 spacer on an Ag substrate is proposed. The dual-band enhanced absorptions in monolayer TMDs are observed, which are related to the SPP and the magnetic resonances. The absorptions of monolayer TMDs are enhanced up to about 70% and 84.5% at the peak wavelengths. The spectral position of the absorption peak can be tuned effectively by changing the period and the radius of the nanodisk array, as well as the thickness of the spacer. In addition, the proposed absorber can tolerate a relatively wide range of incident angles for TE-polarized wave at the second peak. The structure provides an effective way to improve the light-matter interaction for future 2D materials and the resulting applications, such as photodetector and metamaterial absorbers.

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