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Electrical Control of Interband Resonant Nonlinear Optics in Monolayer MoS$_2$

Yunyun Dai,* Yadong Wang, Susobhan Das, Hui Xue, Xueyin Bai, Eero Hulkko, Guangyu Zhang, Xiaoxia Yang, Qing Dai, and Zhipei Sun*

**ABSTRACT:** Monolayer transition-metal dichalcogenides show strong optical nonlinearity with great potential for various emerging applications. Here we demonstrate the gate-tunable interband resonant four-wave mixing and sum-frequency generation in monolayer MoS$_2$. Up to 80% modulation depth in four-wave mixing is achieved when the generated signal is resonant with the A exciton at room temperature, corresponding to an effective third-order optical nonlinearity $\chi^{(3)}_{\text{eff}}$ tuning from ($\sim$12.0 to 5.45) $\times 10^{-18}$ m$^2$/V$^2$. The tunability of the effective second-order optical nonlinearity $\chi^{(2)}_{\text{eff}}$ at 440 nm C-exciton resonance wavelength is also demonstrated from ($\sim$11.6 to 7.40) $\times 10^{-9}$ m/V with sum-frequency generation. Such a large tunability in optical nonlinearities arises from the strong excitonic charging effect in monolayer transition-metal dichalcogenides, which allows for the electrical control of the interband excitonic transitions and thus nonlinear optical responses for future on-chip nonlinear optoelectronics.

**KEYWORDS:** nonlinear optics, four-wave mixing, sum-frequency generation, gate tunability, exciton, MoS$_2$

Atomically thin transition-metal dichalcogenides (TMDs) have stimulated great interest due to their favorable physical properties,$^{1,2}$ such as extremely large exciton binding energy$^3$ and valley pseudospin physics arising from a large spin–orbit interaction.$^4$ In particular, TMD monolayers feature a direct band gap in the visible spectral range,$^{2,5}$ enabling diverse photonic applications including modulators,$^6$ photodetectors,$^7$ and light-emitting diodes.$^8$ Furthermore, TMD monolayers have recently attracted a surge of attention focusing on their fascinating optical nonlinearities$^9$ (e.g., harmonic generation$^{10–12}$ and saturable absorption$^{13,14}$), which are promising for various applications,$^5$ such as wavelength conversion$^{16,17}$ and ultrafast pulse generation.$^{18–20}$

In TMD monolayers, the strongly bound and tunable exciton$^{3,21,22}$ enable the enhanced light–matter interaction$^{3,24}$ and tunable linear and nonlinear optical responses on the atomic thickness scale.$^{25,26}$ For example, it has been demonstrated that the strong interband excitonic effect results in the significant enhancement of various harmonic generation processes$^{27,28}$ in TMD monolayers. Additionally, it has been reported that second-harmonic generation in the WSe$_2$ monolayer can be electrically tuned at A-exciton resonance.$^{26}$ The interband excitonic enhancement and the tunability of nonlinear optical (NLO) responses are fascinating for numerous applications and will enable the versatility of future modern photonics, which is challenging with traditional NLO materials.$^{26,29–31}$ However, the electrical control of the other NLO responses in TMDs still remains unexplored, although it is highly desired for future applications.

In this work, we report the interband excitonic effect of various nonlinear optical processes, including four-wave mixing (FWM) and sum-frequency generation (SFG), in monolayer MoS$_2$ at room temperature. We fabricate an ion-gel-gated monolayer MoS$_2$ device for effective electron doping by applying positive gate voltages. We demonstrate the excitonic enhancement and the gate-tunable FWM of monolayer MoS$_2$. Similarly, for the SFG process, the enhancement and the nonlinear optical tunability at the interband excitonic resonance are also reported. Besides, the second- and third-order nonlinear coefficients are calculated as a function of doping carrier densities.

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RESULTS AND DISCUSSION

Linear Optical Characterization in Monolayer MoS₂. A schematic and an optical image of the MoS₂ device are shown in Figure 1a,b. The device consists of a chemical-vapor-deposited (CVD) monolayer MoS₂ sheet on a SiO₂/Si substrate, with Ti/Au (5/50 nm) as the source and drain electrodes. The ion-gel top-gate method is used for electrostatic gating, which changes the doping level and therefore the optical responses of MoS₂. By examination of the Raman and photoluminescence spectra (Figure S1), the CVD MoS₂ film is identified as a monolayer. Here the electrical property of monolayer MoS₂ is measured with different gate voltages. The transport curve is characteristic of an n-type semiconductor, as shown in Figure 1b. Over a gate voltage range (−3 to 3 V), the source-drain current increases with positive gating (i.e., Vg > 0.4 V), indicating effective electron injection to MoS₂. The low off-current is due to the MoS₂-contact Schottky barrier. Note that in our experimental range (i.e., the gate voltage range is between −3 and 3 V), we are not able to obtain effective hole doping. Therefore, in the following experiments, we mainly study the device at positive gating with effective electron doping.

Figure 1. Monolayer MoS₂ device and its gate-tunable electrical and linear optical properties. (a) Schematic of the gated monolayer MoS₂ device with Ti/Au as source (S) and drain (D) electrodes. Ion gel is used as gate dielectric with Ti/Au as the gate (G) electrode. (b) Source-drain current of the MoS₂ device as a function of the gate voltage Vg from −3 to 3 V with the source-drain bias voltage fixed at 0.1 V. Inset: The optical image of the monolayer MoS₂ channel (white dashed box) with source–drain contacts (yellow regions) on the Si/SiO₂ substrate. Scaler bar: 20 μm. (c) Differential reflection spectra at different gate voltages and the reference linear absorption spectrum (gray) of monolayer MoS₂ at zero gate voltage. A, B, and C excitons are labeled.

Figure 2. Gate-tunable FWM in monolayer MoS₂. (a) Wavelength-dependent FWM spectra. Pump λ₁ is at ~800 nm with an average power of ~1 μW, and idler λ₁ is tunable from ~930 to 1120 nm with an average power of ~1 μW. Inset: Schematic of the FWM of monolayer MoS₂. FWM (ωFWM, red arrow) is generated from monolayer MoS₂ when it is excited by a pump light (ω₁, yellow arrow) and an idler light (ω₂, green arrow). (b) Diagram of the interband resonant FWM process with A(B) excitons. (c) Dependence of FWM peak intensities on incident light powers, with a fit of the experiment data to a power law P. Upper panel: Dependence of FWM on P₁ with fit (s = ~1.90). Lower panel: Dependence of FWM on P₂ with fit (s = ~0.93). (d) Gate-tunable FWM spectra resonant at the A exciton (~650 nm) with λ₁ = 800 nm and λ₂ = 1040 nm. (e) Gate-tunable FWM spectra resonant at the B exciton (~610 nm) with λ₁ = 800 nm, λ₂ = 1160 nm. (f) Normalized FWM peak intensities as a function of gate voltage for FWM resonant at the A- (650 nm, red dots) and B- (610 nm, blue dots) excitonic states and off-resonance (700 nm, black dots).
To understand the excitonic effect on the linear optical properties, the reflection spectra of the MoS₂ device are measured with electron doping at positive gating. The linear absorption spectrum of monolayer MoS₂ on a reference sapphire substrate shows pronounced absorption peaks attributed to A (~650 nm), B (~610 nm), and C (~440 nm) excitonic transitions with no electrostatic gating (gray curve in Figure 1c). However, at a large gate voltage (e.g., $V_g = 3$ V with a doping carrier density of $\sim 4 \times 10^{13}$ cm$^{-2}$), the excitonic effect is strongly suppressed (Figure S2a). The carrier density is estimated using the capacitor model $N = C(V_s - V_{inj})/e$, in which C is the ion-gel capacitance 2.45 $\mu$F/cm$^2$, e is the elementary charge, and $V_{inj} = 0.4$ V is the turn-on voltage in the monolayer MoS₂ device. Here we plot the differential reflection spectra $R(V_g')$ in Figure 1c, where $R(V_g')$ represents the reflection of the monolayer MoS₂ film on the Si/SiO₂ substrate at $V_g'$. The differential reflection spectra clearly show a gate-tunable linear optical response at the A and B excitons. This is mainly because the injected carriers (e.g., electrons when $V_g' > 0.4$ V) decrease the oscillator strength of interband excitonic transitions. The higher the positive gate voltage, the weaker the excitonic-transition-induced linear absorption will be. With a high enough gate voltage (e.g., $V_g = 3$ V), the interband excitonic transitions could even be switched off while a lower energy resonance known as trion (e.g., A-) emerges (Figure S2b). The trion (A-) resonance is quite weak and is observable only by the derivative of the reflection spectra. Therefore, the influence of trion is insignificant in our experiments at room temperature. Note that a negligible change is observed at the C exciton, which is consistent with the result discussed in ref 25.

**Gate Control of Interband Resonant FWM in MoS₂.** A home-built femtosecond-laser-based microscopic system (Figure S3a) is employed for the nonlinear optical measurements in monolayer MoS₂, as described in the Methods section. The monolayer MoS₂ film, excited by two pump frequencies at $\omega_1$ and $\omega_2$ ($\omega_1 > \omega_2$), typically generates various nonlinear optical signals, for example, second-harmonic signals ($2\omega_1$ and $2\omega_2$), SFG ($\omega_1 + \omega_2$), and FWM ($2\omega_1 \pm \omega_2 , \omega_1 \pm 2\omega_2$), as shown in Figure S4.

Here we mainly study the FWM generation (inset of Figure 2a): The pump light ($h\omega_1$) is fixed at ~1.55 eV (i.e., $\lambda_1 = 800$ nm), and the idler light ($h\omega_2$) is tunable in a near-infrared spectral range from ~1.1 to 1.33 eV (i.e., $\lambda_2 = 930$–1120 nm). As a result, the wavelength-dependent FWM ($\omega_{FWM} = 2\omega_1 - \omega_2$) from ~1.77 to 2.00 eV (i.e., $\lambda_{FWM} = 620$–700 nm) is studied, as shown in Figure 2a. The average power for both the pump and idler input light beams in the experiment is ~1 $\mu$W (with a corresponding peak intensity of ~44 GW/cm$^2$). We find that the FWM is enhanced at ~650 nm when the generated FWM photon energy (i.e., $h\omega_{FWM} = 1.91$ eV) matches the A-excitonic energy of monolayer MoS₂, suggesting the interband resonance at the A exciton (Figure 2b).

We further study the interband resonant FWM at the A exciton. Here we use a pump light ($h\omega_1$) at ~1.55 eV (i.e., $\lambda_1 = 800$ nm) and an idler light ($h\omega_2$) at ~1.19 eV (i.e., $\lambda_2 = 1040$ nm), and the generated FWM ($\omega_{FWM} = 2\omega_1 - \omega_2$) is at ~1.91 eV (i.e., $\lambda_{FWM} = 650$ nm). To examine the power dependence of the resonant FWM signal, the nonlinear optical spectra are measured when the power of the pump ($P_1$) and idler ($P_2$) light beams is changed, respectively. The upper panel of Figure 2c shows the peak intensities of the FWM spectra with increasing $P_1$ while $P_2 = 1$ $\mu$W. The lower panel of Figure 2c shows the peak intensities of FWM spectra with increasing $P_2$ while $P_1 = 1$ $\mu$W. It roughly follows a square and linear power-law behavior as a function of $P_1$ and $P_2$, respectively, which confirms the two-color FWM process ($\omega_{FWM} = 2\omega_1 - \omega_2$) involving the pump and idler light beams (details in the Supporting Information). We also study the gate-tunable resonant FWM in monolayer MoS₂ with interband A-excitonic resonance, as shown in Figure 2d. The FWM intensities decrease significantly when $V_g$ changes from 0 to 3 V. We attribute the gate-tunable FWM to the modulation of the oscillator strength of the interband excitonic resonance by the gate-injected electrons. With increasing gate voltages, the oscillator strength at the excitonic resonance decreases with increased doping (as shown in Figure 1c), thus suppressing the exciton enhancement of nonlinear processes. As a result, the FWM signals decrease with increasing gate voltage at the excitonic resonant wavelength. Note that the gate-tunable FWM results are repeatable (Figure S5).

Similarly, with pump light ($h\omega_1$) at ~1.55 eV (i.e., $\lambda_1 = 800$ nm) and idler light ($h\omega_2$) at ~1.07 eV (i.e., $\lambda_2 = 1160$ nm), the generated FWM ($\omega_{FWM} = 2\omega_1 - \omega_2$) at ~2.03 eV (i.e., $\lambda_{FWM} = 610$ nm) is resonant with the B-excitonic state. Figure 2e shows gate-tunable resonant FWM spectra at the B exciton, where the FWM intensities also decrease with increased electrostatic doping due to the same operation principle.
The normalized gate-tunable FWM $\frac{I_{\text{FWM}}(V_g)}{I_{\text{FWM}}(0 \text{ V})}$ is plotted in Figure 2f, where $I_{\text{FWM}}(V_g)$ is the peak intensity of FWM at $V_g$. Here we achieve a nearly 80% (70%) modulation depth $\frac{I_{\text{FWM}}(V_g) - I_{\text{FWM}}(0 \text{ V})}{I_{\text{FWM}}(0 \text{ V})}$ when FWM is resonant at the A(B)-excitonic states with $V_e$ tuning from 0 to 3 V. As a comparison, for the FWM at 700 nm (i.e., $\lambda_1$ at 800 nm and $\lambda_2$ at 920 nm), which is off excitonic resonance, gating does not change the FWM intensity (black dots in Figure 2f). This further demonstrates the dominance of the interband excitonic effect on the optical nonlinearity of monolayer TMDs. Note that the nonlinear coefficients $\chi^{(2)}$ and $\chi^{(3)}$ also use the home-built femtosecond-laser-based microscopic nonlinear intensity, we can estimate the tunability of the excitonic resonances. On the basis of the measured tunable allowing for exceptional control over the oscillator strengths at (Figure 3c), showing a nearly 60% modulation depth $SFG$ at 440 nm decrease with increased electrostatic doping $C$-excitonic resonance. The peak intensities of the resonant excitonic states with generated SFG photon energy (i.e., $\omega_{\text{SFG}} = 2.82 \text{ eV}$), which is due to the fact that the effective electron injection to MoS$_2$ has a turn-on threshold $V_{on}$ at 0.4 V. This critical $V_{on}$ is also indicated by the transport characteristics of monolayer MoS$_2$ (Figure 1b).

**Gate Control of Interband Resonant SFG in MoS$_2$**.

We also use the home-built femtosecond-second-laser-based microscopic system to study SFG in monolayer MoS$_2$ as described in the Methods section. The SFG at the sum frequency ($\omega_{\text{SFG}} = \omega_1 + \omega_2$) is generated with the two input light beams at $\omega_1$ and $\omega_2$. In our SFG experiment, pump1 ($\hbar \omega_0$) is fixed at $\sim 1.55 \text{ eV}$ (i.e., $\lambda_1 \sim 800 \text{ nm}$), and pump2 ($\hbar \omega_0$) is tunable in the near-infrared spectral range from $\sim 1.11$ to 1.33 eV (i.e., $\lambda_2 \sim 930$–1120 nm). As a result, wavelength-dependent SFG is achieved from $\sim 2.66$ to 2.88 eV (i.e., $\omega_{\text{SFG}} \sim 430$–465 nm) (Figure 3a). We find that SFG is enhanced at $\sim 440 \text{ nm}$ when the generated SFG photon energy (i.e., $\omega_{\text{SFG}} \sim 2.82 \text{ eV}$) matches the C-excitonic energy of monolayer MoS$_2$ (inset of Figure 3a). The enhancement suggests that the interband excitonic transition at the C exciton significantly contributes to the enhanced SFG process. The power-dependent SFG signals are examined (Figure 3b). The peak intensities of the SFG spectra scale linearly with pump1 power $P_1$ (while pump2 power $P_2 \approx \sim 1 \mu W$) and $P_2$ (while $P_1 \approx \sim 1 \mu W$), respectively, following the linear power-law behavior of SFG.

We also study the gate-tunable SFG of MoS$_2$ with interband C-excitonic resonance. The peak intensities of the resonant SFG at 440 nm decrease with increased electrostatic doping (Figure 3c), showing a nearly 60% modulation depth $\frac{I_{\text{SFG}}(V_g) - I_{\text{SFG}}(0 \text{ V})}{I_{\text{SFG}}(0 \text{ V})}$ at $V_e = 3 \text{ V}$, where $I_{\text{SFG}}(V_g)$ is the peak intensity of SFG at $V_g$. This demonstrates the gate tunability of the SFG process in monolayer MoS$_2$.

**Gate-Tunable Nonlinear Optical Coefficients**. We have realized the tunability of nonlinear optical signals, which arises from the strong excitonic charging effects in monolayer MoS$_2$, allowing for exceptional control over the oscillator strengths at the excitonic resonances. On the basis of the measured tunable FWM and SFG intensities, we can estimate the tunability of nonlinear coefficients of MoS$_2$. The second- and third-order nonlinear coefficients $\chi^{(2)}$ and $\chi^{(3)}$ are calculated from the measured average powers of the incident light and the generated nonlinear optical signals as follows.

$$\chi^{(2)} = \frac{8n_e}{\alpha_0 \alpha_\phi} \left[ \frac{q_{22} \tau_2 D_2^2 D_1^2 n_1 n_2 e_0 P_1}{\tau_1 D_1^2 n_1 e_0 P_2} \right]$$  

$$\chi^{(3)} = \frac{8n_e}{\alpha_0 \alpha_\phi} \left[ \frac{q_{33} \tau_3 D_3^2 n_2 e_0^2}{P_1} \right]$$  

where $P_{(1,2,3,5)}$ represents the average power of the two incident pump beams and the generated SFG/FWM signals, $f = 2 \text{ kHz}$ is the repetition rate, $\tau_{(1,2,5,5)} = \sim 230 \text{ fs}$ is the estimated pulse duration of the pump light beams and the SFG/FWM signals, $D_{(1,2,5,5)} = \sim 2.5 \mu m$ is the diameter of the laser spot with a Gaussian spatial profile, $n_{(1,2,5,5)} = \sim 4.2$ is refractive index of the material at the pump light frequencies and SFG/FWM frequencies, $\alpha_2 = 2$ for the two-color SFG process, $\alpha_3 = 3$ for the two-color FWM process, and $\psi = (\pi/ln 2)^{1/2}/\pi$. Therefore, the effective bulk-like second- and third-order nonlinear coefficients of monolayer MoS$_2$ ($\chi^{(2)}_{\text{eff}}, \chi^{(3)}_{\text{eff}}$) can be obtained, $\chi^{(2)}_{\text{eff}} = \chi^{(2)}/t$ and $\chi^{(3)}_{\text{eff}} = \chi^{(3)}/t$, where $t = \sim 0.7 \text{ nm}$ is the thickness of the monolayer MoS$_2$. The detailed calibration and calculation method are discussed in the Supporting Information. The typical average power of two pump light beams is $\sim 1 \mu W$ for each light beam. When the gate voltage varies from 0 to 3 V, corresponding to the carrier density change from 0 to $4 \times 10^{13}/\text{cm}^2$, the SFG signal at the C exciton changes from $\sim 2.5$ to 1.0 pW, and the FWM signal changes from $\sim 0.14$ to 0.029 pW at the A exciton and from $\sim 0.093$ to 0.028 pW at the B exciton. Therefore, we obtain the gate-tunable $\chi^{(2)}_{\text{eff}}$ and $\chi^{(3)}_{\text{eff}}$ of monolayer MoS$_2$ as shown in Figure 4.

**CONCLUSIONS**

The interband excitonic resonance of FWM and SFG is investigated in ion-gel-gated monolayer MoS$_2$ at room temperature. The gate-tunable interband excitonic FWM and
SFG are demonstrated by electrostatic doping, which originates from the modulation of the oscillator strength of interband exciton resonances. For interband resonant FWM, $|\chi^{(3)}_{\text{el}}|$ of monolayer MoS$_2$ changes from $(-12.0$ to $5.45) \times 10^{-16}$ m$^3$/V$^2$ at the A exciton (650 nm) and from $(-9.16$ to $5.03) \times 10^{-18}$ m$^3$/V$^2$ at the B exciton (610 nm). Similarly, the tunability of $|\chi^{(2)}_{\text{el}}|$ with the interband C-exciton resonance at 440 nm is tuned from $(-11.6$ to $7.40) \times 10^{-9}$ m/V, with the gate voltage varying from 0 to 3 V. Note that a similar nonlinear optical tunability is anticipated for other TMDs (e.g., WS$_2$). Therefore, our study is promising for electrically controlled NLO devices based on 2D layered materials (e.g., Hamamatsu) in the reflection. Nonlinear optical signals are detected by a spectrometer (Andor, e.g., Poly(styrene)-poly(styrene) (PS) and 0.56 g of ion liquid ([EMIM][TFSI]) in 20 mL of anhydrous dichloromethane). The ion liquid and dry dichloromethane were purchased from Sigma-Aldrich, and PS–PEO–PS was purchased from Polymer Source.

**METHODS**

**MoS$_2$ Device Fabrication.** The monolayer MoS$_2$ is grown on a SiO$_2$/Si substrate by the CVD method.24 The source, drain, and gate electrodes were first patterned using electron-beam lithography (Vistec) and then deposited with Ti/Au (5/50 nm) using electron-beam evaporation. All electrodes were wire-bonded to a chip for electrical control. Ion gel was spin-coated uniformly on the MoS$_2$ device. The ion-gel solution was prepared by dissolving 22 mg of poly(styrene)-b-poly(ethylene oxide)-b-poly(styrene) (PS–PEO–PS) and 0.56 g of ion liquid ([EMIM][TFSI]) in 20 mL of anhydrous dichloromethane.35 The ion liquid and dry dichloromethane were purchased from Sigma-Aldrich, and PS–PEO–PS was purchased from Polymer Source.

**Nonlinear Optical Spectra Measurement.** A home-built femtosecond-laser-based microscopic system is employed for the nonlinear optical experiments. The schematic of this system is shown in Figure S3a. Two incident light beams, with different frequencies, are linearly polarized with polarization directions parallel to each other. Their typical spectra are shown in the Supporting Information. The pulse duration of the incident pulses is $\sim 230$ fs, which is estimated by the cross-correlation measurement (Figure S3c). The pulses of the two pump light beams are spatially and temporally overlapped and focused collinearly on the sample, and they generate the nonlinear optical signals in the monolayer MoS$_2$ film. The spectra of nonlinear optical signals are detected by a spectrometer (Andor, Hamamatsu) in the reflection configuration. The nonlinear optical signals of monolayer MoS$_2$ were measured under different gate voltages. Note that when different source–drain voltages were applied, no change was observed in nonlinear optical signals. Therefore, to fully exclude the electric-current-induced nonlinear optical effects (e.g., current-induced second-order nonlinearity$^{34}$), we only apply gate-voltage and ground source and drain electrodes (i.e., no source–drain current) in our experiments.

**ASSOCIATED CONTENT**

* Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/10.1021/acsnano.0c02642.

MoS$_2$ film characterization, gate-tunable electrical and linear optical properties of monolayer MoS$_2$, FWM and SFG experiment setup and spectra of pump light, nonlinear optical spectra in monolayer MoS$_2$, the repeatable gate-tunable FWM, gate-tunable SFG spectra with excitonic resonance and off-resonance, calculation of nonlinear coefficients for SFG and FWM signals in MoS$_2$, wavelength-dependent third-order nonlinear coefficients of monolayer MoS$_2$, and comparison among different tuning approaches (PDF).

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**Author Contributions**

Y.D. and Z.S. conceived the idea. Y.D. performed the experiments with assistance from Y.W., S.D., H.X., X.B., and E.H. on the device fabrication and characterization. G.Z., X.Y., and Q.D. provided the CVD-grown MoS$_2$ sample. Y.D. analyzed the experimental data. Y.D. and Z.S. wrote the manuscript with contributions from all authors.

**Notes**

The authors declare no competing financial interest.

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