Enhanced optical Kerr nonlinearity of MoS$_2$ on silicon waveguides

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A quasi-two-dimensional layer of MoS$_2$ was placed on top of a silicon optical waveguide to form a MoS$_2$–silicon hybrid structure. Chirped pulse self-phase modulation measurements were carried out to determine the optical Kerr nonlinearity of the structure. The observed increase in the spectral broadening of the optical pulses in the MoS$_2$–silicon waveguide compared with the silicon waveguides indicated that the third-order nonlinear effect in MoS$_2$ is about 2 orders of magnitude larger than that in silicon. The measurements show that MoS$_2$ has an effective optical Kerr coefficient of about 1.1 x 10$^{-16}$ m$^2$/W. This work reveals the potential application of MoS$_2$ to enhance the nonlinearity of hybrid silicon optical devices. © 2015 Chinese Laser Press

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1. INTRODUCTION

Molybdenum disulfide (MoS$_2$) is a kind of transition metal dichalcogenide and has attracted increasing research interest in recent years because of its unique electronic and optical properties [1] that may lead to potential applications in high-performance electronic and optoelectronic devices. Single-layer MoS$_2$ consists of a molybdenum atomic plane sandwiched between two sulphur atomic layers. While bulk MoS$_2$ is an indirect bandgap (about 1.29 eV) semiconductor material, single-layer MoS$_2$ has a direct bandgap of about 1.90 eV [2]. Electrical mobility of 700 cm$^2$/V·s of multilayer exfoliated MoS$_2$ at room temperature has been reported [3], and an extremely high on–off ratio (~10$^6$) field effect transistor was demonstrated with single-layer MoS$_2$ [3,4], which might not be achievable by graphene because of its zero bandgap [5]. Monolayer MoS$_2$ also showed good performance in ultrasensitive photodetectors (PDs) as active materials for optical detection in the ultraviolet to visible range [6]. Moreover, MoS$_2$ was found to be also suitable as a broadband saturable absorber operating at wavelengths in the visible to near-infrared spectral range [7,8]. MoS$_2$ has been used as a saturable absorber in femtosecond mode-locked lasers [7]. Due to the lack of inversion symmetry, odd-layered MoS$_2$ can have second-order nonlinear effects [9], which may be exploited for applications in optoelectronic devices.

Third-order nonlinear effects in MoS$_2$ were previously experimentally studied via the measurement of third-harmonic generation in the wavelength range from 1758 to 1980 nm [10]. The third-order nonlinear susceptibility obtained in that paper was $\chi^{(3)} \sim 10^{-19}$ m$^2$/V at 1758 nm. To our knowledge, however, there has been no previous experimental investigation of the optical nonlinearity of MoS$_2$ at the wavelengths commonly used in telecommunications and silicon photonics. In this paper, a layer of MoS$_2$ prepared by chemical vapor deposition (CVD) was placed on the top of a silicon waveguide integrated with waveguide grating couplers. The interaction of the evanescent optical field in the waveguide with the layer of MoS$_2$ over the length of the waveguide produces changes in the spectra of optical pulses because of self-phase modulation (SPM). The refractive index of MoS$_2$ was obtained experimentally from the redshift of the center wavelength of the grating couplers after placing the MoS$_2$ on the grating coupler. A significant increase in broadening of pulses from the MoS$_2$–silicon waveguide compared to that from the bare silicon waveguide indicates enhancement of the optical Kerr nonlinear effect by the MoS$_2$ layer. Measurement of the spectral broadening also gave estimation of the optical Kerr nonlinear coefficient of MoS$_2$ at a wavelength of 1554.5 nm.

2. DEVICE FABRICATION

The silicon waveguide and grating were fabricated on a silicon-on-insulator wafer with a 340 nm silicon top layer and 2 μm buried oxide. Electron beam lithography (EBL) was used to define the waveguide and grating patterns after a layer of photoresist was formed by spin coating. The chip was developed and rinsed after EBL exposure, followed by etching with deep reactive-ion etching. The MoS$_2$ layer was prepared and transferred onto the silicon chip with methods similar to the techniques described in previous works [11–14]. Solid MoO$_3$ and S powders were used as precursors. Typically, ~50 mg MoO$_3$ (99.5% Sigma Aldrich) was placed in quartz boats and 0.5 g sulphur (99.5% Sigma Aldrich) was located upstream. The CVD growth was performed at atmospheric pressure with 500 sccm (sccm denotes cubic centimeters per minute at standard temperature and pressure) Ar as the protection gas. The growth temperature was around 700 °C.
700°C–800°C. Figure 1(a) shows one set of grating and waveguide, with MoS₂ covering the top of both the grating coupler and waveguide regions. The inset shows the magnified picture for the grating coupler region. Figure 1(b) is the measured Raman spectrum of the MoS₂ layer on the top of the waveguide region.

The effective coupling efficiency of one grating coupler, about 4.5, which is also consistent with the previous report [16]. The loss induced by top-layer MoS₂ could be attributed to the reflection of light by MoS₂ because of the high refractive index. The effective coupling efficiency of one grating coupler, about –6.7 dB, was recorded for determining the input power to the MoS₂–silicon waveguide used in following experiments where third order nonlinear coefficient of MoS₂ will be estimated.

4. SPM MEASUREMENTS AND KERR COEFFICIENT CALCULATION

After transferring MoS₂ onto the silicon waveguide, optical pulses in the waveguide will experience nonlinear SPM from third-order nonlinear effects in silicon and MoS₂, resulting in a broadening of the optical spectrum. Measuring the effective broadened spectrum output from the MoS₂–silicon waveguide and calculating optical mode distribution in the waveguide would allow the third-order nonlinear coefficient of MoS₂ to be obtained. The experimental setup is shown in Fig. 3. Optical pulses at a repetition rate of 1 MHz were produced by gain-switching a distributed feedback (DFB) grating diode laser. The pulses had a full width at half-maximum pulse width of about 56 ps and a center wavelength of 1554.5 nm. The pulses were coupled into the waveguide and the spectral broadening at the output was recorded. A small fraction of the input pulse was directed to the PD for monitoring the average power input to the device under test (DUT). The DUT could be a MoS₂–silicon waveguide, a bare silicon waveguide, or single-mode fiber.

The optical pulses from the gain-switched DFB laser were chirped and the spectrum contains many detailed features, which are difficult to model. Instead of calculating the nonlinear phase shift in the SPM directly, the same spectral broadening as obtained from the MoS₂–silicon waveguide was obtained by SPM in a known length of standard single-mode fiber. The broadened spectrum from the MoS₂–silicon waveguide was first measured, then the DUT was changed to be the single-mode fiber, to which the input power was tuned to make the output spectrum from the fiber match that obtained from the MoS₂–silicon waveguide. With the input power and known parameters of the single-mode fiber, the nonlinear phase shift in the fiber could be calculated, and it is equivalent to that experienced by the pulse in the MoS₂–silicon waveguide. To ensure that the shape of the input pulse spectra to the MoS₂–silicon waveguide and the single-mode fiber were the same, the configuration of the gain-switched laser, erbium-doped fiber amplifier (EDFA) and the bandpass filter (BPF) were unchanged throughout the experiments. The input power to the DUT was adjusted only by a mechanical optical attenuator (ATT). In addition, the fiber between the ATT and the DUT was made as short as possible to avoid spectrum change due to nonlinear effects in this section of fiber. This was verified by measuring the output spectra from the input port to the DUT under various attenuation values, and no significant change in the shape of spectra was found.

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**Fig. 1.** (a) Scanning electron microscope image of one part of the device, with MoS₂ covering both the grating couplers and waveguide regions. As the whole area is covered by the material, significant contrast does not exist. Inset: magnified picture of the grating. (b) Measured Raman spectrum of the MoS₂ layer on the top of the waveguide region.

**Fig. 2.** (a) Experimental results of the transmission spectra of the gratings. Significant redshift of the center wavelengths was induced by the MoS₂ layer. (b) FDTD simulation result for the redshift of the grating to estimate the refractive index of MoS₂.

**Fig. 3.** Experimental setup used for measuring the third-order nonlinear coefficient of MoS₂. The gain-switched DFB diode laser was used as pulse source. PC, polarization controller.
The output spectra from the MoS₂–silicon waveguide and single-mode fiber were then measured and recorded with the optical spectrum analyzer (OSA). The results are shown in Fig. 4. In Fig. 4, the black curve shows the input spectrum, while the red curve and the blue curve correspond to broadened output spectra from the MoS₂–silicon waveguide and the bare silicon waveguide, respectively. Evidently, the pulse broadening effect is more significant in the MoS₂–silicon waveguide, which indicates a larger nonlinear Kerr coefficient of the MoS₂ layer. The calculation of the coefficient is carried out as follows.

Nonlinear phase shift experienced by the pulse in MoS₂–silicon waveguide is given by [17,18]

\[ \phi^{\text{MOS}}_{\text{nl}} = \frac{2\pi \Delta n_{\text{eff}} L_{\text{eff}}}{\lambda}, \tag{1} \]

where \( \Delta n_{\text{eff}} \) is effective nonlinear refractive index of the MoS₂–silicon waveguide, \( \lambda \) is the vacuum center wavelength of the pulse, and \( L_{\text{eff}} = [1 - \exp(-\alpha L)]/\alpha \) is the effective length of the structure (\( L \) corresponds to the physical length). Change in effective index due to the nonlinear optical Kerr effect could be treated as a small perturbation. According to the first-order perturbation theory [19], \( \Delta n_{\text{eff}} \) can be expressed as

\[ \Delta n_{\text{eff}} = \frac{\int_{0}^{\infty} \Delta \epsilon_r |E|^2 \, dx \, dy}{2Z_0 \int_{0}^{\infty} \text{Re} \{ E^* \times H \} \cdot \hat{e}_z \, dx \, dy}, \tag{2} \]

where \( \Delta \epsilon_r \) is the change of dielectric constant, and \( Z_0 \) is the impedance of the vacuum. The pulse is assumed to propagate along the \( z \) direction. The integral in Eq. (2) covers the whole cross section of the structure. Denoting the Kerr coefficient as \( n_2 \), the change of nonlinear Kerr constant can then be approximated by

\[ \epsilon_r = \left( n + \Delta n \right)^2 \approx n^2 + 2n \Delta n, \]

\[ \Delta \epsilon_r \approx 2n \Delta n = n^2 n_2 c_0 |E|^2, \tag{3} \]

where \( n \) is refractive index, \( \Delta n = n_2 I \) is the small change of refractive index due to the Kerr effect, \( I = \frac{1}{2} c_0 n |E|^2 \) is local optical intensity, \( c_0 \) is vacuum permittivity, and \( c \) is the speed of light in vacuum space.

For the high index contrast waveguide, \( H \neq \frac{\omega}{c} \hat{e}_z \times E \). Therefore, the denominator in Eq. (2), which is \( \int_{0}^{\infty} \text{Re} \{ E^* \times H \} \cdot \hat{e}_z \, dx \, dy \) cannot be simplified as the conventional method

[20]. Fortunately, from the relationship of power flux and energy density flow [20], we have

\[ \frac{1}{2} \int_{-\infty}^{\infty} \text{Re} \{ \hat{H}^* \cdot \hat{E} \} \cdot \hat{e}_z \, dx \, dy = \frac{1}{2} \frac{v_g}{v_p} \int_{-\infty}^{\infty} |\hat{E}|^2 \, dx \, dy, \tag{4} \]

where \( v_p \) is group velocity for the pulse in the waveguide and \( \epsilon = c_0 n^2 \). With Eqs. (1)–(4), one can finally get that

\[ \Delta n_{\text{eff}} = \frac{n_g^2 P_0}{A_{\text{eff}}^2} \sum_n n_2 A \int_{-\infty}^{\infty} |\hat{E}|^4 \, dx \, dy, \tag{5} \]

where \( n_2 \) is group index; \( A \) denotes the MoS₂, silicon, and silicon dioxide regions, and \( A_{\text{eff}} \) is defined as

\[ A_{\text{eff}} = \frac{\left( \int_{-\infty}^{\infty} |\hat{E}|^2 \, dx \, dy \right)^2}{\epsilon_0 \int_{-\infty}^{\infty} |\hat{E}|^4 \, dx \, dy}. \tag{6} \]

Integration of the amplitude of the electric field can be obtained by mode distribution over the MoS₂–silicon structure from the simulation, which is shown in Fig. 5.

Nonlinear phase shift of the single-mode fiber can be calculated with the following equation [18]:

\[ \phi^{\text{Fiber}}_{\text{nl}} = \frac{2\pi n_2 \frac{P_0 L_{\text{eff}}}{\lambda}}{A_{\text{eff}}}, \tag{7} \]

where \( n_2 \) is the Kerr coefficient of the fiber, \( P_0 \) the peak power input to the fiber, and \( A_{\text{eff}} \) the mode effective area, which could also be calculated with the mode distribution from simulation for the fiber. In this paper, a piece of 102 m standard single-mode fiber was used, whose linear loss is \( \sim 0.4 \text{ dB/km} \), and \( n_2 \gamma \sim 2.25 \times 10^{-20} \text{ m}^2/\text{W} \) [18]. \( P_0 \) was adjusted by ATT in Fig. 3, so as to make the output spectrum from the fiber match that of the MoS₂–silicon waveguide, so that

\[ \phi^{\text{MOS}}_{\text{nl}} = \phi^{\text{Fiber}}_{\text{nl}}. \tag{8} \]

With this method and Eqs. (1)–(8), the Kerr coefficient of MoS₂ was extracted to be about \( 1.1 \times 10^{-16} \text{ m}^2/\text{W} \), with a measurement uncertainty around 40%. The uncertainty comes from several aspects. First, the coupling efficiency per grating was estimated from the total insertion loss (IL) of the waveguide at low input power. The efficiency is modeled to follow normal distribution whose mean is half of the IL, and the 3σ region covers half of the IL. So the actual input power to the

Fig. 4. Input and output spectrum of MoS₂–silicon waveguide and bare silicon waveguide. Black curve is the input spectrum, while the red and blue curves are from the output of the MoS₂–silicon waveguide and the bare silicon waveguide, respectively.

Fig. 5. Optical intensity distribution of the MoS₂–silicon waveguide structure, which was used for nonlinear refractive index calculation. MoS₂ layer, silicon core, and silicon dioxide substrate are indicated.
MoS$_2$–silicon waveguide accounts for a large uncertainty factor. Second, the Kerr coefficient of silicon $n_{2\text{Si}}$ was used in calculating the coefficient for MoS$_2$ [Eq. (4)], and it also has a range of uncertainty based on previous measured results [21], $n_{2\text{Si}} = 4.5 \times 10^{-18}$ m$^2$/W is applied here. Third, the equivalent nonlinear phase shift determined from the fiber also has uncertainty in matched input power to the fiber, accounting for the uncertainty in determining the nonlinear phase shift. These three factors together are responsible for the overall uncertainty of around 40%. Nevertheless, the deduced coefficient can provide a reference for the order of magnitude. It is 2 orders larger than that of silicon, and from the equation [22]

$$n_2 = \frac{3}{4\epsilon_0cn^2} \chi^{(3)},$$

(9)

where $n$ is the refractive of MoS$_2$, which is about 4.5 from the result above, the third-order susceptibility $\chi^{(3)}$ of MoS$_2$ is thus about $7.9 \times 10^{-18}$ m$^2$/V. The result is 1 order of magnitude larger than the value measured at 1758 nm in work by Wang et al. [10].

5. CONCLUSIONS

In conclusion, a quasi-two-dimensional sheet of MoS$_2$ was transferred on top of a silicon waveguide and the optical parameters were measured at telecommunication wavelength. The refractive index was extracted to be $\sim 4.5$ from the redshift of the grating couplers. The third-order nonlinear coefficient and susceptibility of MoS$_2$ was determined from SPM in the MoS$_2$–silicon structure. The nonlinear refractive index of the material was extracted to be about $1.1 \times 10^{-16}$ m$^2$/W, and the corresponding susceptibility is about $7.9 \times 10^{-18}$ m$^2$/V. The high nonlinear coefficient of MoS$_2$ at telecommunication wavelengths offers opportunities to use it to enhance the optical nonlinearity of hybrid silicon photonic waveguides for nonlinear optical devices.

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