Research article

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Broadband spatial self-phase modulation and ultrafast response of MXene $\text{Ti}_3\text{C}_2\text{T}_x$ ($\text{T}=\text{O}$, OH or F)

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Abstract: Two-dimensional layered materials (2DLM) have become the subject of intensive research in various applications such as electronics, photonics and optoelectronics due to their unique physical properties. As a new class of 2DLM, MXenes have attracted great interest due to their superior performance in a wide variety of applications such as batteries, supercapacitors, catalysts, electronics and optics. Here, we have investigated the broadband spatial self-phase modulation (SSPM) and ultrafast response of the MXene $\text{Ti}_3\text{C}_2\text{T}_x$ ($\text{T}=\text{O}$, OH or F) experimentally. The MXene $\text{Ti}_3\text{C}_2\text{T}_x$ exhibited the broadband nonlinear optical response via SSPM from 400 nm to $\sim 1\ \mu\text{m}$ under the ultrafast laser excitation, and ultrafast carrier characteristics with an ultrafast recovery time with femtosecond transient absorption spectroscopy. The experimental results have shown that the MXenes have the broadband nonlinear optical response, which can lay a foundation for the application prospect for the MXene-based ultrafast optoelectronic devices.

Keywords: nonlinear optics; ultrafast dynamics; spatial self-phase modulation; MXene.

1 Introduction

Two-dimensional layered materials (2DLM) possess unique physical and chemical performance for easy-to-assemble building blocks for nanoscale structures, which have fascinated various novel applications ranging from optical communication to optical sensing, etc. Starting with the 2DLM graphene, different 2D materials have been investigated and applied in various fields, such as metal oxides and hydroxides, topological insulators, disulfides and hexagonal boron nitride [1–9]. In addition, novel 2D materials have been prepared and investigated with specific requirements and technical evolvments. In 2011, Naguib et al. [10] put forward a new class of 2D transition metal carbides, nitrides, and carbonitrides which were labeled as MXenes for they were produced by selectively etching the A layer from their three-dimensional (3D) layered parent compound MA_nX_n or MAX, where M is an early transition metal (such as Ti, Mo, Sc, Zr, Hf, V, Cr, Nb and Ta), A includes Al or Si, X is carbon and/or nitrogen, and n is 1–3 [11, 12]. The multilayer MXenes including $\text{Ti}_3\text{C}_2\text{T}_x$, Ti_2CT_x , Mo_2CT_x , V_2CT_x , Nb_2CT_x , $\text{Nb}_4\text{C}_3\text{T}_x$, $\text{Ta}_4\text{C}_3\text{T}_x$ and so on have been synthesized [13–16]. In this representation, T_x represents surface groups, mainly including oxygen ($-\text{O}$), fluorine ($-\text{F}$), or hydroxyl ($-\text{OH}$) [10, 17]. MXenes consist of stacked 2D nanosheets that combine with weak van der Waals forces and/or hydrogen bonds between these surface groups, providing the possibility of layering them into few-layer MXene colloidal solution, of which $\text{Ti}_3\text{C}_2\text{T}_x$ has been widely studied. MXenes are nearly metallic with high electron density near the Fermi level (E_F), a unique combination of metal conductivity and hydrophilicity, high specific capacitance and good cycle stability, and excellent mechanical flexibility. Excellent performance makes MXenes ideal electrode materials for supercapacitors, lithium batteries and ion batteries [16, 18–23]. Furthermore, they have great application potential in energy storage, microwave absorption, electronics, optics and so on [14, 24–27].

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When it comes to the optics regime, MXenes have shown excellent optical response for their strong interaction with light. Jiang et al. systematically studied the broadband nonlinear optical properties of $\text{Ti}_3\text{C}_2\text{T}_x$ in the infrared wavelength region [28]. Based on the nonlinear saturable absorption characteristics of MXenes, they have been adopted in the field of ultrafast pulsed lasers towards the mid-infrared spectral range [29–32]. Meanwhile, the MXene $\text{Ti}_3\text{C}_2\text{T}_x$ and C_{60} (reverse saturable absorber) were stacked in series to manufacture passive photonic diodes to realize the irreversible transmission of light, which is equivalent to the application of isolators in fiber-based lasers [33]. In addition, based on the high thermal conductivity of MXenes, all-optical modulators, loaders, and switches were designed in space or micro-fiber due to their outstanding photothermal effect [34–36]. Although the nonlinear optical response of the MXenes and optoelectronic applications have been investigated, the ultrafast carrier response of the MXenes has been less explored, which is important and urgent for nonlinear optical applications.

Spatial self-phase modulation (SSPM), a coherent third-order nonlinear optical effect initially observed in the nematic liquid crystal in 1981 [37], has developed as a typical method to investigate the nonlinear optical response of the low-dimensional materials [38–46]. The refractive index of a Kerr medium is a quantity that is related to the intensity of the incident light, and the nonlinear optical susceptibility $\chi^{(3)}$ is uniquely determined by the laser-intensity-dependent refractive index $n = n_0 + n_2 I$, where n_0 and n_2 are linear and nonlinear refractive indexes, respectively [38]. When the incident laser passes through the sample, the optical phase depends on the transverse light intensity distribution, which will result in a conical outgoing diffraction and interference in the far field, i.e. SSPM diffraction ring. The diffraction ring directly reflects the optical phase and is correlated to the electronic phase and coherence of the material [38].

Here, we have investigated the broadband nonlinear optical response of the MXene $\text{Ti}_3\text{C}_2\text{T}_x$ via SSPM, Z-scan and femtosecond transient absorption spectroscopy experimentally. We found that the MXene has the carrier characteristics with an ultrashort intraband carrier recovery time of ~ 0.2 ps and broadband nonlinear optical properties. Inspired by the narrow bandgap of the MXene, we carried out a broadband SSPM experiment to study its nonlinear optical properties [37, 47], and the experimental results show that $\text{Ti}_3\text{C}_2\text{T}_x$ exhibited broadband nonlinear absorption and a large nonlinear refractive index. The experimental results suggest that the MXene

is an ultrafast and broadband nonlinear optical material that can be used to design and fabricate novel nonlinear optical devices.

2 Preparation and characterization of $\text{Ti}_3\text{C}_2\text{T}_x$

The MXene $\text{Ti}_3\text{C}_2\text{T}_x$ was prepared by etching, which is based on the method reported by Naguib et al. [10]. Ti_3AlC_2 powders ($\geq 98\%$ purity; Beijing Lianlixin Technology Co., Ltd., Beijing, China) were soaked in an aqueous hydrogen fluoride (HF) solution (40%) for 2 days at 25°C . By washing several times with deionized water, the generating precipitate was vacuum-dried at 60°C for 2 days. The accordion-like structure of the layered $\text{Ti}_3\text{C}_2\text{T}_x$ after etching can be presented through a scanning electron microscope (SEM), as shown in Figure 1A and B. In addition, we can find that the flakes have different orientations. The crystal features of MXene $\text{Ti}_3\text{C}_2\text{T}_x$ nanosheets were characterized by transmission electron microscopy (TEM), as shown in Figure 1C and D. The interlayer distance is confirmed as $9.84 \text{ \AA}$, which is close to the previous reported value of $9.93 \text{ \AA}$ [28], as shown in Figure 1F. The atomic lattice under high-resolution transmission electron microscopy (HRTEM) and the selected area electron diffraction (SAED) image of $\text{Ti}_3\text{C}_2\text{T}_x$ are well characterized in Figure 1E and G. In Figure 1H, Raman spectra measured by an inVia Reinshaw confocal spectrometer (wavelength: 633 nm ; power: $60 \text{ \mu W}$; Reinshaw, Gloucestershire, UK) has main bands centered at 270 , 403 and 612 cm^{-1} , corresponding to the Ti–C bond vibration, which showed the expected vibrational modes for the MXene $\text{Ti}_3\text{C}_2\text{T}_x$ as the previous reports [10, 48, 49]. The linear absorption characteristics of $\text{Ti}_3\text{C}_2\text{T}_x$ have been characterized by a spectrophotometer UV-3600 Plus (Shimadzu, Kyoto, Japan), which shows that $\text{Ti}_3\text{C}_2\text{T}_x$ has a broadband absorption, as shown in Figure 1I.

3 Nonlinear optical properties of the MXene $\text{Ti}_3\text{C}_2\text{T}_x$

3.1 Broadband SSPM experiments

The broadband SSPM experiments have been performed to validate the nonlinear optical response of the MXene, as shown in Figure 2. Ultrafast pulsed lasers with the wavelengths of 800 nm and 1064 nm were

Figure 1: Material characteristics of the MXene $\text{Ti}_3\text{C}_2\text{T}_x$.

(A, B) SEM images of delaminated $\text{Ti}_3\text{C}_2\text{T}_x$ after HF etching. (C, D) TEM images of $\text{Ti}_3\text{C}_2\text{T}_x$ flakes. (E) Atomic lattice of $\text{Ti}_3\text{C}_2\text{T}_x$ under an HRTEM. (F) The interlayer distance is measured to be 9.84 Å, close to the theoretical value of 9.93 Å [28]. (G) SAED image of $\text{Ti}_3\text{C}_2\text{T}_x$. (H) Raman spectrum of $\text{Ti}_3\text{C}_2\text{T}_x$ after HF etching. (I) Linear absorption spectrum of $\text{Ti}_3\text{C}_2\text{T}_x$.

Figure 2: Broadband SSPM experiments.

(A) The experimental setup diagram of the SSPM nonlinear optical experiment. (B) Stable diffraction rings at 400 nm and (C) 800 nm wavelengths. (D) Snapshots of the pattern formation under 1064 nm wavelength.

used as experimental light sources, respectively, and a 400 nm wavelength light can be obtained by inserting a frequency doubling crystal into the optical path

of the 800 nm femtosecond pulsed path. As shown in Figure 2, the prepared MXene $\text{Ti}_3\text{C}_2\text{T}_x$ dispersion loading in a quartz cuvette was placed on the focus of light for

experiments. After the focused laser beam interacted with the MXene dispersion, the SSPM effect occurred with a diffraction ring in the far field. In the experiment, diffraction ring images appeared at wavelengths of 400 nm, 800 nm and 1064 nm, respectively, as shown in Figure 2. The snapshots in Figure 2D show pattern formation of the SSPM effect at 1064 nm wavelength. The phenomenon of expansion and distortion of the diffraction rings over time can be clearly captured at all experimental wavelengths. The intensity distributions of the experimental results in Figure 2 and the corresponding theoretical simulation results were shown in the Supplementary material.

Based on the optics Kerr effect, the nonlinear refractive index of $\text{Ti}_3\text{C}_2\text{T}_x$ can be expressed as follows:

$$n = n_0 + n_2 I \quad (1)$$

where n_0 is the linear refractive index of the $\text{Ti}_3\text{C}_2\text{T}_x$ dispersion, n_2 is the nonlinear refractive index of the MXene $\text{Ti}_3\text{C}_2\text{T}_x$ and I is the intensity of the incident light. When the light beam is focused and passed through the $\text{Ti}_3\text{C}_2\text{T}_x$ dispersion, the SSPM effect begins to form and the phase shift can be expressed as follows [37]:

$$\Delta\Psi = \frac{2\pi n_0}{\lambda} \int_0^{L_{\text{eff}}} n_2 I(r, z) dz \quad (2)$$

where λ is the wavelength of light, L_{eff} is the effective optical propagation length and $I(r, z)$ is the intensity distribution. The L_{eff} of $\text{Ti}_3\text{C}_2\text{T}_x$ can be obtained by [42]

$$L_{\text{eff}} = \int_{L_1}^{L_2} \left(1 + \frac{z^2}{z_0^2} \right)^{-1} dz = z_0 \arctan \left(\frac{z}{z_0} \right) \Big|_{L_1}^{L_2} \quad (3)$$

where z_0 is the Rayleigh length, z is the propagation length, L_2 or L_1 stands for the distance between the front surface or back surface of the cuvette and light focus, and L is the thickness of the sample. The phase shift of a Gaussian beam can be expressed as follows [43]:

$$\Delta\Psi = \Delta\Psi_0 \exp(-2r^2/a^2) \quad (4)$$

where a is equal to $1/e^2$ beam radius. The bright and dark rings alternately occur when the number of rings and the phase shift satisfy the relationship $\Delta\Psi(0) - \Delta\Psi(\infty) = 2N\pi$. After the derivations, the nonlinear refractive index of $\text{Ti}_3\text{C}_2\text{T}_x$ can be written as

$$|n_{2, \text{total}}| = \frac{\lambda}{2n_0 L_{\text{eff}}} \cdot \frac{N}{I} \quad (5)$$

The third-order nonlinear susceptibility $\chi^{(3)}$ is an important parameter of nonlinear optics material, and the effective nonlinear refractive index can be expressed as [43]

$$|n_2| = \left(\frac{12\pi^2}{n_0^2 c} \right) 10^3 |\chi^{(3)}| \quad (6)$$

The third-order nonlinear susceptibility of $\text{Ti}_3\text{C}_2\text{T}_x$ is defined as $\chi_{\text{total}}^{(3)}$. From the slope of the ring number N and light intensity I , $\chi_{\text{total}}^{(3)}$ of $\text{Ti}_3\text{C}_2\text{T}_x$ can be written as

$$|\chi_{\text{total}}^{(3)}| = \frac{c\lambda n_0}{2.4 \times 10^4 \pi^2 L_{\text{eff}}} \cdot \frac{dN}{dI} \quad (7)$$

The third-order nonlinear susceptibility of monolayer $\text{Ti}_3\text{C}_2\text{T}_x$ nanosheets can be obtained by

$$|\chi_{\text{total}}^{(3)}| = |\chi_{\text{monolayer}}^{(3)}| N_{\text{eff}}^2 \quad (8)$$

where N_{eff} is the effective number of the monolayer $\text{Ti}_3\text{C}_2\text{T}_x$ nanosheets in dispersion, and $n_{2, \text{monolayer}}$ can be obtained in the same way.

Figure 3 demonstrates the linear relationship between the ring number and different light intensity under two laser sources. After fitting, we can obtain the nonlinear refractive index $|n_{2, \text{total}}|$ and the third-order nonlinear susceptibility $|\chi_{\text{total}}^{(3)}|$ at 800 nm and 1064 nm wavelengths, respectively. The effective number N_{eff} of layered $\text{Ti}_3\text{C}_2\text{T}_x$

nanosheets can be obtained from $T_{\text{monolayer}}^{N_{\text{eff}}} = T_{\text{total}}$, where

$T_{\text{monolayer}}$ is the transmittance of a $\text{Ti}_3\text{C}_2\text{T}_x$ monolayer, which was 96.6% [34]. T_{total} is the transmittance of the $\text{Ti}_3\text{C}_2\text{T}_x$ dispersion, as shown in Figure 1D. The transmittance was about 10%; then the effective number of $\text{Ti}_3\text{C}_2\text{T}_x$ monolayers is calculated to be ~66. The third-order nonlinear optical susceptibility ($|\chi_{\text{monolayer}}^{(3)}|$) of the monolayer $\text{Ti}_3\text{C}_2\text{T}_x$ is $\sim 10^{-15}$ ($|n_{2, \text{monolayer}}| \sim 10^{-18}$) at 800 nm and $\sim 10^{-7}$ ($|n_{2, \text{monolayer}}| \sim 10^{-10}$) at 1064 nm wavelength. The conclusion is closer to the results of the Z-scan study, as shown in the Supplementary material.

We have summarized the results of the related SSPM experiment in Table 1. By comparing these reported results, it can be seen that graphene has a larger third-order nonlinear polarizability $\chi^{(3)}$, mainly due to its Dirac cone structure. For the nearly metallic properties of the MXene $\text{Ti}_3\text{C}_2\text{T}_x$, we obtained a nonlinear polarizability $\chi^{(3)}$ of 10^{-7} (e.s.u.) at 1064 nm wavelength, which has almost the same magnitude as graphene. It can be known from Equation 7 that $\chi^{(3)}$ is mainly related to the ring number N and the laser intensity I . In our experiments, the difference from $\chi^{(3)}$ in the experiments is mainly due to the different laser

Figure 3: The relationship between N and I under different laser illumination. (A) 800 nm and (B) 1064 nm wavelength ultrafast lasers.

Table 1: Third-order optical susceptibilities of the reported materials.

Sample	Laser parameter	$\chi^{(3)}_{\text{monolayer}}$	Ref.
MoS ₂	532 nm CW laser, 400 nm and 800 nm ultrafast lasers	10^{-9} (e.s.u.) at visible to near-infrared wavelengths	[38]
MoSe ₂	488 nm CW laser	10^{-9} (e.s.u.)	[40]
Graphene	532 nm TEM ₀₀ -mode CW laser	10^{-7} (e.s.u.)	[43]
Black phosphorous	350–1160 nm femtosecond lasers	10^{-8} (e.s.u.) at multiple wavelengths	[44]
Graphite	532 nm CW laser	2.2×10^{-9} (e.s.u.)	[46]
MoTe ₂	473 nm, 532 nm, 750 nm, 801 nm CW lasers	10^{-9} (e.s.u.)	[50]
Ti ₃ C ₂ T _x	800 nm femtosecond laser and 1064 nm picosecond laser	10^{-15} (e.s.u.) at 800 nm and $\sim 10^{-7}$ (e.s.u.) at 1064 nm	This work

parameters. In addition, the MXene flakes with different orientations may make different effects during the interaction between the MXene and light, such as the absorption and scattering process. However, it is a collective behavior caused by all flakes during the dynamic process before the diffraction ring stabilizes in the SSPM experiment. We will try to design and observe the effect of the orientation of the flakes in the subsequent experiments.

In a previous experiment, the diffraction ring exhibited a collapse distortion in the vertical direction of light path; the asymmetric thermal convection introduced by gravity played a major role [45, 51]. As shown in Figure 4C, the Gaussian beam first expanded to maximize diffraction rings at 0.4 s after the sample was irradiated. Subsequently, the mode of the circular diffraction rings distorted to organize a semicircular distortion ring within ~ 1.2 s because of the non-axisymmetric thermal convection of the MXene Ti₃C₂T_x induced by light. On the basis of the previous experiment, the sample was placed horizontally on a higher level of the light path and the laser beam was vertically focused upwards to study the influence of gravity, as shown in Figure 4A. Comparatively, when

Figure 4: The SSPM experiments. (A) Schematic diagram of the SSPM experimental setup with the sample placed horizontally. (B, C) Snapshots of the pattern formation at 800 nm wavelength.

the laser beam focused on the sample vertically, the ring reached its maximum and steady state within ~ 1.2 s with no distortion, as shown in Figure 4B. This result indicates

that the asymmetric thermal convection in the SSPM effect is mainly due to the gravity effect.

3.2 Light intensity-dependent collapse

The SSPM effect shows intensity-dependent collapse behavior with the increasing intensity of light. The schematic of the traditional SSPM phenomenon is given in Figure 5, and the extent of the collapse can be described by the change of diffraction angle formed by the sample [39]. When the SSPM effect appears, the diffraction ring forms a series of coaxial cones behind the sample. The distortion angle can be described as

$$\theta_D = \theta_H - \theta'_H = \frac{R_H}{D} - \frac{R'_H}{D} = \frac{R_D}{D} \quad (9)$$

where R_H is the maximum diffraction radius, θ_H is the maximum half diffraction angle, and θ_D can also be displayed as the following expression [37, 47]:

$$\theta_D = \theta_H - \theta'_H = (n_2 - n'_2)IC = \Delta n_2 IC \quad (10)$$

Finally, via removing the distortion maximum half angle θ'_H at the stable state, the relationship between the nonlinear refractive index variable and the maximum half angle can be obtained following the expression

$$\frac{\Delta n_2}{n_2} = \theta_D / \theta_H \quad (11)$$

We have measured the distance from the CCD to the sample and recorded the size of the diffraction ring. The change of diffraction angles with the increase of light intensity at 800 nm and 1064 nm wavelengths can be obtained, and the change of n_2 can be calculated using the above equation, as shown in Figure 5. As the laser intensity increases, a larger nonlinear phase shift can be obtained, which results in a larger radius and more ring numbers. On the other hand, a larger light intensity results in stronger thermal convection, resulting in a lower density in the upper portion of the MXene

Figure 5: Light intensity-dependent collapse in SSPM experiments.

(A) Image and schematic of diffraction ring distortion at 800 nm wavelength. The tendency of the half cone angle (B, E), collapse angle (C, F) and $\Delta n_2/n_2$ (D, G) as the increases of the intensity of 800 nm and 1064 nm wavelengths, respectively.

dispersion. Therefore, the change of maximum diffraction half angle θ_H and distortion angle θ_D are almost linearly proportional to the incident intensity. Although the change in n_2 increases with the increasing intensity due to strong thermal convection, it tends to be saturated at higher light intensity.

4 Ultrafast carrier characteristics

To further understand the light-MXene interaction, we studied the ultrafast carrier characteristics of the MXene $\text{Ti}_3\text{C}_2\text{T}_x$ by femtosecond transient absorption spectroscopy [52]. During the measurement, 90% of the output beam of the laser (center wavelength: 400 nm; pulse duration: 190 fs; repetition rate: 6 kHz) was used as the pump light to excite the sample with an energy of 1.5 μJ . Ten percent of the output beam passed through a time delay device and focused onto the nonlinear crystal (sapphire) to generate the supercontinuum white light as the probe light, which was focused into the sample after collimation and filtered to remove the fundamental frequency light. After the sample was modulated, the signal carrying the information of the sample was collected and recorded by a linear array photodiode in the grating spectrometer. By adjusting the time delay line, the carrier dynamics of the sample can be characterized, as shown in Figure 6. The change of optical density with time delay at a single wavelength can be obtained by $\Delta\text{OD} = -\lg(T/T_0)$, where T is the transmittance of the sample under pumping and T_0 is the linear transmittance of the sample. In Figure 6A, the

more prominent positive signal indicates that the excited state absorption dominates after the sample was excited. From the transient absorption spectrum, the kinetic curve closer to the infrared wavelength was extracted and fitted, and two processes with lifetimes of ~ 0.2 ps and ~ 4 ps were obtained, as shown in Figure 6B. The initial ultrashort carrier recovery time constant (0.2 ps) corresponds to the intraband thermal scattering of the excited carriers and reaches thermal equilibrium, while the slower time constant (4 ps) is attributed to the interaction of hot carriers-phonons and cooling, and the recombination time of the electrons-holes [53–56]. The ultrafast carrier dynamics characteristic makes MXenes promising nonlinear optical materials for ultrafast applications.

Figure 7 can help us to understand the light-MXene interaction in the SSPM experiment. Similar to the case of the liquid crystal, the optical nonlinearity can be put down to the reorientation and alignment of $\text{Ti}_3\text{C}_2\text{T}_x$ nanosheets caused by the electromagnetic field [37, 38, 43, 57]. The reorientation here resulted from the motion of electrons introduced by the oscillating electric field of the laser. For the mechanism of the light-MXene interaction, electrons in the valence band can be excited to the conduction band by absorbing incident photons, and then they will relax back to the bottom of the conduction band, which mainly contains two stages for the recovery process. The carrier-carrier scattering keeps the first few hundred femtoseconds, and the carrier-phonon relaxation plays the leading role in the next few picoseconds until the electrons stabilize. Based on the narrow bandgap characteristics of $\text{Ti}_3\text{C}_2\text{T}_x$ (the dotted lines represent the band structure caused by the difference of surface groups), photons with excitation wavelengths

Figure 6: Ultrafast dynamic process of the MXene $\text{Ti}_3\text{C}_2\text{T}_x$. (A) Transient absorption curve and (B) carrier dynamic curve of $\text{Ti}_3\text{C}_2\text{T}_x$.

Figure 7: The mechanism of the interaction between light and MXene $\text{Ti}_3\text{C}_2\text{T}_x$.

Inset: Gaussian nonlinear phase shift; the coherence of wave vectors with the same phase (such as positions r_1 and r_2) results in the formation of the diffraction ring [37, 38, 43].

of 800 nm and 1064 nm can provide energy to help carriers perform the transition. During the interaction process, the electrons move in the opposite direction to holes along the electric field when the laser is present, thus producing the polarized $\text{Ti}_3\text{C}_2\text{T}_x$ nanosheets. The reorientation and alignment of $\text{Ti}_3\text{C}_2\text{T}_x$ nanosheets caused the minimal interaction with the electromagnetic field. The change of nonlinear refractive index will result in a diffraction ring, and each nanosheet contributes constructively to the SSPM ring pattern.

5 Conclusions

In summary, the high-quality MXene $\text{Ti}_3\text{C}_2\text{T}_x$ nanosheets were synthesized by the HF etching method. The ultrafast carrier characteristics with an intraband recovery time of ~ 0.2 ps and larger nonlinearity make MXenes excellent candidates for ultrafast optoelectronic devices. The SSPM effect of the MXene $\text{Ti}_3\text{C}_2\text{T}_x$ was observed from visible to near-infrared (400–1064 nm), highlighting its broadband nonlinear optical properties. By studying the SSPM effect with the excitation of ultrafast pulsed lasers, the large third-order nonlinear susceptibility $\chi^{(3)}$ of the MXene $\text{Ti}_3\text{C}_2\text{T}_x$ was extracted. In addition, the gravity effect results in an asymmetric thermal convection of the MXene $\text{Ti}_3\text{C}_2\text{T}_x$

dispersion, leading to a collapse of the diffraction pattern. Our work confirmed that the MXene $\text{Ti}_3\text{C}_2\text{T}_x$ is an ideal broadband nonlinear optical material, which shows great potential for nonlinear ultrafast optoelectronic devices.

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