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# Effect of concentration on the formation time of diffraction rings in spatial self-phase modulation

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A new unsaturated wind-chime model is proposed for calculating the formation time of the diffraction rings induced by spatial self-phase modulation (SSPM) in molybdenum disulfide suspension. To optimize the traditional wind-chime model, the concentration variable of 2D materials was introduced. The results of the unsaturated wind-chime model match quite well with the SSPM experimental results of molybdenum disulfide. Based on this model, the shortest formation time of diffraction rings and their corresponding concentration and light intensity can be predicted using limited data. Theoretically, by increasing the viscosity coefficient of the solution, the response time of the diffraction ring, to reach the maximum value, can be significantly reduced. It has advanced significance in shortening the response time of photonic diodes.

**Keywords:** spatial self-phase modulation; wind-chime model; nonlinear optics. **DOI:** 10.3788/COL202220.011901

#### 1. Introduction

It has been 40 years since spatial self-phase modulation (SSPM) was discovered in 1981<sup>[1]</sup>. Since then, SSPM has developed rapidly due to the simple experimental device required to produce it, wide measurable wavelength range, strong diffracted beam, and feasibility for materials with a relatively large gap<sup>[2]</sup>. The research materials include a large number of two-dimensional (2D) materials, such as Bi<sub>2</sub>TeSe<sub>2</sub><sup>[3]</sup>, graphene<sup>[4,5]</sup>, molybdenum disulfide<sup>[6–8]</sup>, MXenes<sup>[9–11]</sup>, and other<sup>[12–19]</sup>. Tremendous effort has been put into comprehending the underlying mechanism for the appearance of the SSPM patterns. The theoretical reports involve several factors such as concentration<sup>[20]</sup>, light intensity<sup>[21]</sup>, and thickness of a cuvette<sup>[22]</sup>. With the development of nonlinear optics<sup>[23–26]</sup>, today SSPM has become a ubiquitous, convenient, and efficient method for characterizing the third-order nonlinear optical properties of 2D materials<sup>[27,28]</sup>.

Recently, all-optical switching and information converters based on SSPM have been developed<sup>[29]</sup>. A nonreciprocal light propagation device has also been proposed based on the graphdiyne/SnS<sub>2</sub> structure<sup>[30]</sup>. Furthermore, taking advantage of the strong light–matter interaction, a hybrid dimensional heterojunction-based logic gate is designed to realize the OR function with the optical method<sup>[31]</sup>. The successful preparation of graphdiyne-PMMA film<sup>[30]</sup> and the Te@Bi quantum dots (QDs) nanotubes (NTs) PMMA film<sup>[31]</sup> significantly promoted the progress of self-diffraction in photoelectric applications.

Nevertheless, photoelectric applications are inseparable from the key parameter of response time, but hardly ever reported.

The "wind-chime" model is an ideal model proposed by Zhao's group based on non-local electronic coherence theory, which can be used to calculate the formation time of the diffraction rings from the initial to maximum number of rings. When the light is incident on the sample, the nanosheets begin to be polarized. Initially, there is an arbitrary angle between the nanosheet and the electric field. Thanks to energy relaxation, the electric field reorients the nanosheets so that each domain contains an axis parallel to the polarization direction of the external field, just like a "wind chime"<sup>[2]</sup>. An unsaturated wind-chime model is put forward in this work by introducing the concentration variable to satisfy different concentration conditions. These findings will no doubt promote the theoretical development of SSPM.

#### 2. Materials and Methods

The  $MoS_2$  nanosheets were synthesized via conventional liquid phase exfoliation<sup>[32]</sup>. As a result, the size of the nanosheets is difficult to be uniformly perfect. Figure 1(a) shows the scanning electron microscope (SEM) image of  $MoS_2$ . The nanosheets show an average size with hundreds of nanometers, which will induce typical SSPM diffraction rings. In previous reports, the results show that the size of the nanosheets will influence the nonlinear optical response when the size changes from hundreds of nanometers nanosheets to several nanometers QDs<sup>[20]</sup>. Here, the size changes within a smaller range than that of the previous report. So, we mainly explored the influence of the concentration on the formation time of the diffraction rings without considering the size dependent effect of SSPM. Scanning transmission electron microscopy (STEM) and energy-dispersive X-ray spectroscopy (EDX) mapping images for MoS<sub>2</sub> are presented in Fig. 1(b). The Mo and S elements show overlapped morphology structure, which confirmed the elemental composition of the as-prepared MoS<sub>2</sub>. The oxygen element can also be observed due to unavoidable oxidation during the preparation of the samples. An atomic force microscope (AFM) measurement was performed to characterize the thickness of the MoS<sub>2</sub> nanosheets, and the results are shown in Figs. 1(c) and 1(d). The average thickness of MoS<sub>2</sub> nanosheets is about 5.3 nm.

The experimental setup of SSPM is shown in Fig. 2(a). A femtosecond (fs) laser regenerative amplifier from Spectra-Physics was utilized as the experimental light source. The laser is incident at a wavelength of 700 nm (it can be seen from Ref. [18] that at different wavelengths, the formation time of the diffraction rings is nearly the same) with a repetition frequency of 2 kHz and a pulse width of about 40 fs. Due to the use of the fs laser, the heat generation is very limited during the nonlinear optical response measurement here. The focal length of the lens was 20 cm, and the cuvette size was 1 cm × 1 cm × 4 cm. While testing MoS<sub>2</sub>, its front surface was 10 cm away from the lens, and the back surface was 22.5 cm from the laser beam profiler (LBP2-VIS2). The camera features a resolution array of 964 × 724, the acquisition mode is 12 bits, and the time resolution is up to 30 frames per second.

### 3. Results and Discussion

When light passed through the sample, diffraction rings began to appear until the radius of the SSPM pattern reached its maximum [Figs. 2(b1)-2(b4)]. Then, the upper part of the diffraction rings collapsed downwards to form a stable conical structure that is vertically asymmetric [Fig. 2(b5)], which is caused by the thermal convection phenomenon<sup>[4]</sup>.

Furthermore, the reorientation of a 2D material can be explained according to the following process: under laser irradiation, photo-excited electrons and holes move in opposite directions, which polarizes the 2D material. The polarized 2D material would be reoriented parallel to the electric field to minimize the system's energy. The formation time of diffraction rings in Figs. 2(b1)-2(b4) is equal to the time required for the nanosheets reorientation in Figs. 2(c2)-2(c3). Moreover, the ring formation time *T* can be calculated as follows<sup>[2]</sup>:

$$T = \frac{\varepsilon_r \pi \eta \xi r c}{1.72(\varepsilon_r - 1)Ih},\tag{1}$$

where *r* is the radius of the fragment, *h* is the thickness,  $\xi$  is a compensation value,  $\varepsilon_r$  is the permittivity,  $\eta$  is the solvent's viscosity, and *I* is the laser intensity. However, the change in the sample concentration was not considered in this model. In this work, the improvement for different sample concentrations based on this model is proposed.

In the traditional model<sup>[2]</sup>, all nanosheets are considered to participate in the wind-chime model and contribute to the number of rings. Our previous work demonstrated that not all



**Fig. 1.** (a) SEM image of  $MoS_2$  nanosheets. (b) STEM image and EDX mapping images of  $MoS_2$  nanosheets. (c) AFM image of  $MoS_2$ . (d) The height profiles corresponding to the solid lines in (c).



**Fig. 2.** (a) Schematic setup of SSPM. (b1)–(b5) The dynamic process of the diffraction ring patterns. (c1)–(c3) Unsaturated wind-chime model changes with the increase in time.

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fragments are involved in forming the wind-chime model<sup>[20]</sup>. In other words, the number of rings is related only to the number of nanosheets participating in the wind-chime model.

In fact, in the general "wind-chime" model, the total number of fragments has been determined once the sample solution is determined<sup>[20]</sup>:

$$A_{\text{total}} = \frac{\rho R^2 l}{r^2 h},\tag{2}$$

where *R* is the radius of the beam waist,  $\rho$  is the volume concentration of the solution, and *l* is the effective optical path of the laser through the sample:

$$l = \int_{l_1}^{l_2} \left( 1 + \frac{z^2}{z_0^2} \right)^{-1} dz, \qquad z_0 = \frac{\pi \omega_0^2}{\lambda}.$$
 (3)

 $l_1$  and  $l_2$  are the distances from the front surface and the back surface of the cuvette to the lens, respectively. The number of effective nanosheets is related to the total number of fragments and the incident light intensity  $I_0$  within the effective length l. The proportion of active flakes,  $P_{\text{total}}$ , is a function of the incident light intensity I and the effective optical path l <sup>[20]</sup>:

$$A_{\rm eff} = A_{\rm total}(l,\rho) \times P_{\rm total}(I,l).$$
(4)

At a fixed concentration, the number of SSPM rings is proportional to the number of fragments M, which have already generated electron coherence. The light intensity determines the number of fragments that can generate electron coherence. Still, it is not inversely proportional to M, because the change in light intensity changes the number of coherent fragments and the number of SSPM rings simultaneously (at different intensities, producing one ring needs different M). Therefore, we introduced the proportionality coefficient  $\chi$ . Finally, it is organized into

$$M = \frac{N}{I}\chi.$$
 (5)

Moreover, according to the optical Kerr effect, the nonlinear refractive index  $n_2$  can be described as<sup>[33]</sup>

$$n_2 = \frac{\lambda}{2n_0 l} \cdot \frac{N}{I},\tag{6}$$

where  $\lambda$  is the excitation wavelength, and  $n_0$  is the linear refractive index. As can be seen in Eq. (6), the proportionality coefficient  $\chi$  is related to parameters of the sample, such as the nonlinear refractive index.

In the ideal model, the fragments in the solution are evenly distributed from 0 to  $\pi/2$ . At different concentrations, the average rotational angle of fragments that generated electron coherence  $\varphi$  should be proportional to the effective amount of fragments *M*. Therefore, the following equation can be obtained:

$$\frac{\varphi}{\pi/2} = \frac{M}{\rho \pi R^2 l / \pi r^2 h},\tag{7}$$

which finally turns into<sup>[2]</sup>

$$\varphi = \frac{(\varepsilon_r - 1)}{\varepsilon_r} \frac{2 t I h}{4 \eta \xi r c} \cdot N \cdot \overline{\sin 2\theta_i}, \qquad (8)$$

$$T = t \cdot N, \tag{9}$$

$$T = \frac{n\pi\chi\varepsilon_r\eta\xi r^3c}{0.86(\varepsilon_r - 1)I^2R^2l\rho}.$$
 (10)

As can be seen from Fig. 3(a), the number of SSPM rings keeps increasing constantly as the concentration remains relatively small. As the concentration reaches the saturation value at  $0.2 \text{ cm}^3/L$  or so, the number of rings no longer increases. In Fig. 3(b), the formation time of diffraction rings decreases with increasing concentration. It is concluded that specific light intensity can make a limited number of MoS<sub>2</sub> fragments produce electronic coherence. When the light intensity is saturated, the SSPM rings can no longer change even if the concentration



Fig. 3. (a) Number of rings versus different concentrations. (b) The experimental values and two kinds of theoretical values of ring formation time at different concentrations.

increases. There are three concentrations corresponding to the number of rings of seven because the concentration has a small effect on the number of rings<sup>[20]</sup>.

In Fig. 3(b), the black points represent the experimental values, the red dots represent the theoretical values calculated by Eq. (10), and the blue dots represent the theoretical values calculated by Eq. (1). Through experiments, we found that the formation time will decrease as the concentration increases under the same light intensity. Especially at high concentrations  $(0.52 \text{ cm}^3/\text{L})$ , the theoretical result of the original formula (1.66 s) is about eight times the experimental value (0.21 s).

It is difficult for the original formula to calculate the formation time of the diffraction rings precisely under different concentrations.

Comparing the theoretical and experimental values, it is found that the theoretical value can also show the trend of the change in theoretical data such as  $0.26 \text{ cm}^3/\text{L}$  and  $0.032 \text{ cm}^3/\text{L}$ , which indirectly verifies the credibility of the formula presented in this work. The error of the data largely comes from lowconcentration samples because there are fewer 2D fragments. When the sample concentration is low, the proportion of thermal effects increases, and the nonlinear optical characteristics are weakened. Since the thermal effect is not considered in this article, there is a difference between the experimental data and the calculated data.

Equation (10) can be used to accurately fit the relationship between the formation time and the sample concentration at the same light intensity. The formation time of the diffraction rings is related to the sample concentration and closely related to the incident light intensity. Using this formula, the experimental values for the different incident light intensities are compared with the theoretical values under the condition that MoS<sub>2</sub> concentration is consistent with the value of  $\chi$  in the calculation process. Figure 4 displays the comparison of the theoretically calculated value and experimental value at the concentrations



Fig. 4. Time varying with light intensity and concentration. The cubic dots represent the experimental values, and the spherical dots represent the theoretical values.

of  $0.52 \text{ cm}^3/\text{L}$ ,  $0.26 \text{ cm}^3/\text{L}$ , and  $0.065 \text{ cm}^3/\text{L}$ . At high concentration, the difference between the theoretical and experimental values is within the error range (0–0.14 s). With the decrease of light intensity at low concentration, the gap between the theoretical value and the experimental value becomes larger and larger and is gradually distorted.

#### 4. Conclusions

Combining all of the above results, we found that the formation time of the diffraction rings gradually decreases with the increase in light intensity and the increase in the concentration. The surface fitted by a small amount of experimental data can predict the formation time of the diffraction rings under specific concentrations and intensities. In other words, it provides a facile way to find the condition of concentration and light intensity corresponding to the shortest formation time. Due to the limitation of our detector's resolution, we cannot observe a response time less than 0.07 s. There are saturation values for both light intensity and concentration, and hence from Eq. (10), the response time cannot be infinitesimally small. Besides, due to the complexity of the formula, the formation time cannot become infinitely small by only changing the concentration. But, the viscosity coefficient can be infinitely large. In other words, the reorientation of fragments is eliminated by preparing solid films. In this way, the number of rings will not increase over time, and the response time will be significantly reduced. The method we proposed is theoretically feasible, though it can be complicated to get diffraction ring patterns in solid materials.

Using the experimental data of  $MoS_2$  at different concentrations, a new unsaturated wind-chime model was successfully established. By comparison, the calculated data is consistent with the experimental data, verifying the feasibility of the formula at different materials and different concentrations. At the same time, the formula is also applicable for calculating the ring formation time of the 2D layered material at different intensities. Finally, the shortest formation time of diffraction rings can be predicted by limited experimental data. The results obtained herein have advanced significance for photonic devices such as photodiodes.

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