Determination of the waist location for the Gaussian beam based on second harmonic generation of monolayer MoS$_2$

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Abstract. The waist location of the Gaussian beam was determined based on nonlinear responses of monolayer MoS$_2$ on a SiO$_2$/Si substrate. Monolayer MoS$_2$ was characterized by the Raman spectroscopy, whose nonlinear spectrum was measured and whose second harmonic generation (SHG) was detected obviously. While monolayer MoS$_2$ was moved along z axis, its SHG intensity decreased after increasing and the location with maximum SHG was the Gaussian beam waist location. The change of the SHG intensity at the two sides of the beam waist was symmetrical. Our finding is beneficial to the calculation of the waist radius and the optical property research of MoS$_2$ and some electromagnetic materials furtherly.

1. Introduction

Two dimensional (2D) nanomaterials have attracted more and more attention after a discovery of graphene [1,2], it is due to their extremely thin thickness, unique morphology, and excellent performance. There are many kinds of 2D nanomaterials, including graphene, BN, transitional metal sulphides (TMDS) [3-7], and MoS$_2$ is most concerned in TMDS [8-15]. Besides some fields including circuits [16,17], electronics [18-21] and optoelectronics and photonics [22,23], nonlinear optical properties of MoS$_2$ capture great interests of many researchers currently. Owing to broken inversion symmetry, distinct second-order nonlinearity exhibits in a MoS$_2$ layer with odd layer, especially single layer [24]. Some researchers have explored second harmonic generation (SHG) of monolayer MoS$_2$ at present. Kumar et al. and Malard et al. observed highly significant SHG in monolayer MoS$_2$ [25,26]. Li et al. detected SHG of MoS$_2$ and h-BN whose thickness were from monolayer to five layers [27]. Clark et al. probed the polycrystalline property of monolayer MoS$_2$ grown through chemical vapor deposition [28]. Le et al. also observed strong SHG of MoS$_{2(1-x)Se_{2x}}$ with single layer by Se doping [29].

Key parameters of the Gaussian beam are the waist location, the waist size and the beam divergence angle, especially the waist location and the waist size who can determine transmission characteristics of Gaussian beam entirely, and the determination of the waist location for the Gaussian beam was studied at the very beginning of the laser era, in 1960s and 1970s of the last century. In practice, it is necessary to know specific values of the waist location and waist size in order to meet some special requirements, such as the design of the laser beam transmission or the imaging system, the study of Gaussian beam transmission characteristics, the study of nonlinear optics and the application of industrial or medical lasers. There are many specific methods for measuring Gaussian beam parameters, such as the over-coring method, the know-edge method and the CCD method [30-34]. For the over-coring method and the knife-edge method, their characteristics are to obtain a power
or intensity distribution by measuring the power or intensity of a beam at a certain location in different range, figure out the beam radius at these locations, and derive the waist location and waist radius based on the free transmission characteristics of a Gaussian beam reversely, but the measurement is slow, and the change of the light intensity during the measurement process can cause measuring errors easily. In addition, the wavefront distortion of the Gaussian beam also brings some errors for the measurement of the beam radius, and the radius of the beam is also involved in the calculation during the derivating process, which makes the measurement result having large error. For the CCD method, its characteristic is to capture the entire beam’s contour and provide a 2D distribution of the beam’s intensity simultaneously, so that the beam radius can be obtained and the waist location and waist radius can be deduced reversely according to the free transmission characteristics of Gaussian beam. This measuring method is not affected by the change of the light intensity, the measuring speed is fast, but the size of a pixel becomes a factor of the measuring accuracy and the high resolution beam quality analyzer is expensive. In addition, the wavefront distortion of Gaussian beam in practice will also result in a large error in measuring process. In this study, we detected the SHG of monolayer MoS2, which is a concerned nanomaterials in recent years. It was found that the SHG intensity of monolayer MoS2 changed gradually when MoS2 was moved along z axis. We determined the waist position (or focus position) of the Gaussian beam based on the SHG intensity change of monolayer MoS2. In the process, we can not only measure the nonlinear properties, but also determine the waist position by the way.

2. Experimental section

MoS2 films were mechanically exfoliated on 300 nm SiO2/Si substrate. The colors of the MoS2 films were observed through a microscope. Raman spectra of MoS2 films were determined by a Raman spectrometer (Invia, Renishaw) with 514 nm laser line. A femtosecond (fs) oscillator (Mira 900S, Coherent) generated a 800 nm fs laser light with a 76 MHz repetitive frequency and a 130 fs pulse width, and the fs laser light was focused on MoS2 films by a 60× objective len (NA = 0.85) equipped at an inverted microscope (Axio Observer A1, Zeiss). Nonlinear optical signals of MoS2 films were gathered by the same objective len and conducted to a composition of a spectrometer (SR-500i-B1, Andor) and a coupled-charge device (DU970N, Andor) for the exploration, as shown in Fig. 1. For moving MoS2 layers along Z axis, MoS2 was put on a three dimensional located system (P-563.3CD, Physik Instruments) with a 1 nm accuracy, the direction of X axis and Z axis is shown in the right-upper corner of Fig. 1.

3. Results and discussion

3.1. Principle for measuring Gaussian beam waist location

The measuring principle for the Gaussian beam may be explained through Fig. 2. The laser beam, launched from the laser, becomes a Gaussian beam to be measured (Incident Beam) after passing through the attenuator (M) and the initial focus lens (LensⅠ), the waist radius of the incident beam is $R_0$, the distance of the waist location away from LensⅠ is $L_0$. The waist radius of the incident beam who is focused is $R_0'$, the distance of the waist location away from LensⅡ is $L_0'$. The CCD camera is
fixed on the digital display moving table, in practice, the table can change the position of the CCD camera.

For Gaussian beam ordinarily, the change of the beam radius near the beam waist is very small and its value is close to that of the waist radius, so it is difficult to measure the waist radius and determine the waist location directly and precisely. However, while the Gaussian beam travels away from the waist location, the beam will diverge and the beam radius increases and changes significantly, so that beam radius can be measured at these positions accurately, even with ordinary measuring instruments, such as a low-cost CCD camera. Since the Gaussian beam is symmetrical relative to the beam waist, in practice, if beam radiuses of \( z_1 \) and \( z_2 \) with equal values are measured shown in Fig. 2, and the number of the beam radius is \( R_{z_1} \) and \( R_{z_2} \), and \( R_{z_1} = R_{z_2} \), the middle point of \( z_1 \) and \( z_2 \) is the Gaussian beam waist location \( L_0 \) which can be expressed as

\[
L_0 = z_1 + \frac{z_2 - z_1}{2}
\]

According to the formula (1), the waist location can be determined.

![Fig. 2 Measuring setup for Gaussian beam.](image)

Because the beam intensity at two cross sections with equal radiuses is equal for the Gaussian beam, nonlinear response of nanomaterials in the two cross sections is equal due to the laser excitation. Therefore, the intensity of nonlinear response for nanomaterials at both sides of beam waist is also symmetrical. Referring the formula (1), we can determine the waist location through utilizing the symmetry of nonlinear responses. Our measurement for Gaussian beam based on Fig. 1 is similar to Fig. 2. In this study, the SHG of monolayer MoS2 exfoliated on a 300 nm SiO2/Si substrate was detected. It was found that SHG intensities of monolayer MoS2 changed gradually when MoS2 was moved along Z axis. We determined the waist location of the Gaussian beam based on the SHG intensity change of monolayer MoS2.

3.2. Characterization of monolayer MoS2 on a SiO2/Si substrate
We first detected linear optical properties of the MoS2 film whose microscope image was shown in the inset of Fig. 3, the thicknesses of MoS2 films were characterized by Raman spectra. Raman spectra of monolayer MoS2 were measured at 514 nm laser line, as shown in Fig. 3. Two peaks for the in-plane (E\(_{2g}\)) and out-of-plane (A\(_{1g}\)) vibration modes were distinctly exhibited, whose positions were 384.5 cm\(^{-1}\) and 403.1 cm\(^{-1}\) separately. The MoS2 film was demonstrated to be monolayer because the Raman shift difference between E\(_{2g}\) and A\(_{1g}\) modes is 18.6 cm\(^{-1}\) [35].
3.3. Nonlinear optical response spectra measurements

Since monolayer MoS$_2$ was with a large $\chi^{(2)}$ [25-29], we measured nonlinear response spectra for monolayer MoS$_2$ on the SiO$_2$/Si substrate. The excitation power dependence of nonlinear response spectra was shown in Fig. 4. The inset exhibits the change of SHG intensities on excitation powers drew in a dual logarithmic coordinate. The linear fitting gives a slope of ~1.96, perfectly matching the second order property of the nonlinear optical process.

3.4. Measurement of the waist location based on nonlinear optical responses of monolayer MoS$_2$ moved along z axis

In order to determine the waist location of the excitation beam, we measured nonlinear spectra of monolayer MoS$_2$ moved along z axis in Fig. 1. The plane with the largest SHG was set as $z = 0$, $z$ was positive above the plane and $z$ was negative below the plane. Nonlinear spectra, which were measured through moving monolayer MoS$_2$ along z axis every 2.5 µm, are shown in Fig. 5(a). SHG intensity changes of monolayer MoS$_2$, which was moved along z axis every 0.5 µm, are exhibited in Fig. 5(b), the result indicates that SHG intensities decrease after increasing. Because the largest laser intensity appears at the Gaussian-beam waist plane, the location with strongest SHG is the waist location. Moving the sample, SHG of monolayer MoS$_2$ increases with the enhancement of the excitation beam intensity. Then moving to the waist location, SHG reaches the maximum. Continue to move the sample which will be away from the waist position, the excitation beam intensity gradually weakens, and SHG weakens gradually with the decrescence of the beam intensity. The change of SHG at both sides of the beam waist is symmetrical. If the plane with the largest SHG is not set as $z = 0$, the coordinates of the beam waist location with the largest SHG can be defined on the basis of formula (1). Only now the SHG intensity of $z_1$ and $z_2$ possess equal values, do not refer to beam radius, and $z$-
axis direction is vertical shown in Fig. 1. Therefore, the waist location can be determined by observing where the maximum value of SHG of monolayer MoS$_2$ happened.

When every other parameter of the laser remains known, the waist size and spatial mode can be inferred after the determination of the waist location. Because MoS$_2$ and some electromagnetic materials can show good optical properties at the waist location, the determination of the waist location is advantageous for their study. Certainly, the technique can just be generalized to measure the waist sizes of pulsed laser, as only intense lasers show significant SHG. Because of the successful application of MoS$_2$ at the waist position measurement, it can be inferred that other nanomaterials with good nonlinear properties can also be used to achieve this application, such as ZnO nanorods, Si quantum dots. In addition, the presented method doesn’t fully achieve the determination of the waist size, only determinates the waist location and isn’t even cheap, but the method shows the possibility of the laser-parameter determination by nonlinear optical properties of nanomaterials.

4. Conclusion
In summary, we determined the waist location of the Gaussian beam based on the symmetrical change of the SHG intensity at both sides of the beam waist for monolayer MoS$_2$ on the SiO$_2$/Si substrate. Monolayer MoS$_2$ was characterized by the Raman spectroscopy whose nonlinear spectrum was measured and whose SHG was detected obviously. While monolayer MoS$_2$ was moved along z axis, SHG intensity of monolayer MoS$_2$ decreased after increasing, and the location with maximum SHG was the Gaussian beam waist location. Our finding is helpful to the calculation of waist radius, the optical properties research of MoS$_2$ and some electromagnetic materials furtherly.

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