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# A method for measuring the continuous dispersion spectrum of nonlinear refraction coefficients of materials

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ABSTRACT

In this paper, a method for measuring the continuous dispersion spectrum of nonlinear refraction coefficients of materials based on complex refractive index dispersion (CRID) measurement is proposed. This method involves measuring internal reflection spectra with a lab-made apparatus. From the determined CRIDs of a material with and without exciting light, the changes in refractive index over a wide spectral range can be obtained, from which the nonlinear refraction coefficients can be deduced. In addition, the RI changes at wavelengths far from the nonlinear range can be used to monitor the thermal effects. In this study, a methyl-red-doped poly(methyl methacrylate) (MR-PMMA) sample was investigated. A large nonlinear refraction coefficient on the order of $10^{-1}$ $cm^2/W$ was observed. The results also show that the absolute value of nonlinear refraction coefficient of the MR-PMMA sample decreases with the increase of excitation intensity. Consequently, this study provides a powerful approach that has the potential to be applied in the study of the nonlinear properties of materials.



## 1. Introduction

The relationships between the optical properties of materials and external conditions, such as temperature [1,2], electric intensity [3], physiological status [4,5] and exciting light intensity [6–16], have drawn the interests of many researchers. The thermo-optical coefficient and electro-optical coefficient are very important parameters when the optical material is applied in thermal or electric environment. In biomedical field, the optical property changes can reflect the oxygen content in blood, or the physiological status of tissue. Optical parameters, including nonlinear absorption and nonlinear refraction, are very important in nonlinear optical systems.

The nonlinear refraction has been widely studied owing to its potential application in nonlinear optical devices. The commonly used third order nonlinear refraction coefficient, defined as $n_2$ ($cm^2/W$), is an important parameter to characterize optical nonlinear material. Theoretically, $n_2$ can be calculated from the equation $n_2 = \Delta n_r / I$, where $\Delta n_r$ is the refractive index (RI) change and $I$ represents the excitation light intensity [6,7]. Four-wave mixing [8] and self-diffraction [9] are important nonlinear phenomenons that can be applied to investigate the nonlinear refraction coefficient, but the setup is complicated. The Z-scan method proposed by Sheik et al. [6] is a means of determining $n_2$ by utilizing the self-focusing and self-defocusing phenomena. Even though the Z-scan method is unsuitable for the determination of nonlinear refraction over a wide spectral range, its simple structure and straightforward procedure make it the most widely used nonlinear refraction research approach. Various kinds of materials have been investigated by the Z-scan method, such as azobenzene-molecule-doped materials [7] [10–12], bacteriorhodopsin (BR) film [13], and graphene [14]. The light intensity at the focal point of the Z-scan beam is very high, so the thermal effects should be taken into account during Z-scan measurement. Song et al. [13] observed that the nonlinear refraction of BR film changed from positive to negative nonlinear phenomenon with the increase of light intensity. Recently, pulsed lasers have become the most commonly used light sources in the Z-scan experiment due to their low thermal effects [12,14,15]. However, even when pulsed lasers are used, thermal effects will occur if the pulse repetition time is improper [15]. Actually, the thermal effects can also be potentially applied in optical-limiting devices [2]. Nevertheless, if the objective is to determine nonlinear refraction coefficients, the thermal effects must be eliminated or

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well defined in the analysis. Besides that, the nonlinear optical phenomenon is always complicated, which is far complicated than that the third order coefficient can describe.

When $\Delta n_r$ is measured at the same wavelength as exciting light, $n_2$ is a self-modulation coefficient. In contrast, $n_2$ is a cross-modulation coefficient when $\Delta n_r$ is measured at other wavelengths. Both self-modulation and cross-modulation are important, so a method that can determine the optical parameters over a continuous spectral range is required. The white-light interferometry method has been applied to measure electro-optical coefficients over continuous spectral range [3], but it is not suitable for absorptive materials. Therefore, a method, which is applicable for various kinds of materials, is needed.

The complex refractive index (CRI), defined as $CRI = n_r(1 + i\kappa)$, contains the real RI $n_r$ and the extinction coefficient $\kappa$ [17]. For absorptive material, the values of $n_r$, $\kappa$ and the absorption coefficient $\alpha$ satisfy $\alpha = 4\pi n_r \kappa/\lambda$, where $\lambda$ represents the wavelength. Thus, CRI changes induced by exciting light contain both nonlinear refraction and nonlinear absorption information. In this study, we utilized a previously proposed lab-made apparatus [18,19] to measure the CRI dispersion (CRID) of methyl-red-doped poly(methyl methacrylate) (MR-PMMA) sample under different excitation intensities and to determine its nonlinear refraction coefficients. Several new results were observed.

## 2. Materials and methods

### 2.1. Materials

Methyl-red-doped poly(methyl methacrylate) (MR-PMMA), was chosen as a representative material. MR is a kind of azobenzene molecule, and its nonlinear optical properties have been studied by various researchers. To prepare the sample, MR powder and PMMA in a mass ratio of 3:97 were dissolved in cyclohexanone liquid. Then, the mixed liquid was poured onto the surface of the prism. The MR-PMMA sample was obtained when the cyclohexanone completely evaporated.

### 2.2. Experimental apparatus

A top-down view of the experimental apparatus is illustrated in Fig. 1. The apparatus consists two parts. The first part is the reflection measurement part which is similar to that in Refs. [18,19]. In this part, four arms, labeled $L_1$, $L_2$, $L_3$, and $L_4$, are connected by high-precision bearings with intersection points $J_1$, $J_2$, $J_3$, and $J_4$. The lengths of $L_1$ and $L_2$ are equal, so are those of $L_3$ and $L_4$. The point $J_1$ can move along the $J_1J_2$ direction driven by a stepper motor. $J_2$ coincides with the center of the semi-cylindrical prism, and $J_1J_2$ is perpendicular to the prism-sample interface. Arm $L_3$ is used to mount a light source fiber, a beam expander, and an aperture. The probing beam from arm $L_3$ is first reflected by the prism-sample interface, then passes through a concave lens ($f$ = -50 mm), and finally reaches the fiber spectrometer (HR4000, Ocean Optics). Geometrically, the incident angle $\theta$ can be deduced from Eq. (1).

$$\theta = \arccos[(b^2 + s^2 - a^2)/(2bs)], \tag{1}$$

where $b$, $s$, and $a$ are the lengths of $J_1J_3$, $J_1J_2$, and $J_1J_3$, respectively. With the apparatus, reflection spectra at various incident angles can be measured. Refractive index of water from Ref. [20] is used to calibrate the apparatus, and previous study on BK7 glass shows that the discrepancies between measured refractive index dispersion and theoretical values are less than 0.0008 [18].

The second part is the excitation part. 532 nm continuous laser light (P-polarized) propagates through a beam-expander, an aperture (diameter: 2 mm) and then perpendicularly irradiates on the prism-sample interface.

### 2.3. Complex refractive index dispersion determination

In this study, each reflection measurement is performed twice. One is for sample and the other is for air. The reflection intensity ratio of sample and air is the light reflectance of the sample. In total, there are 1353 wavelengths in 400–750 nm and reflectance curve at each wavelength contains 250 incident angles. At a fixed wavelength $\lambda$, a multi-curve fitting method based on Nelder-Mead algorithm [21] is adopted to deduce the CRI of material $n$ by minimizing the sum

$$S(N) = \sum_{1}^{j_{\max}} (R_{m,j} - R_{C,j})^2 \tag{2}$$

where $j$ is the number of incident angle, $R_{m,j}$ and $R_{C,j}$ are the measured reflectance and the calculated reflectance based on Fresnel equations [17]. For $S$-polarized and $P$-polarized light,

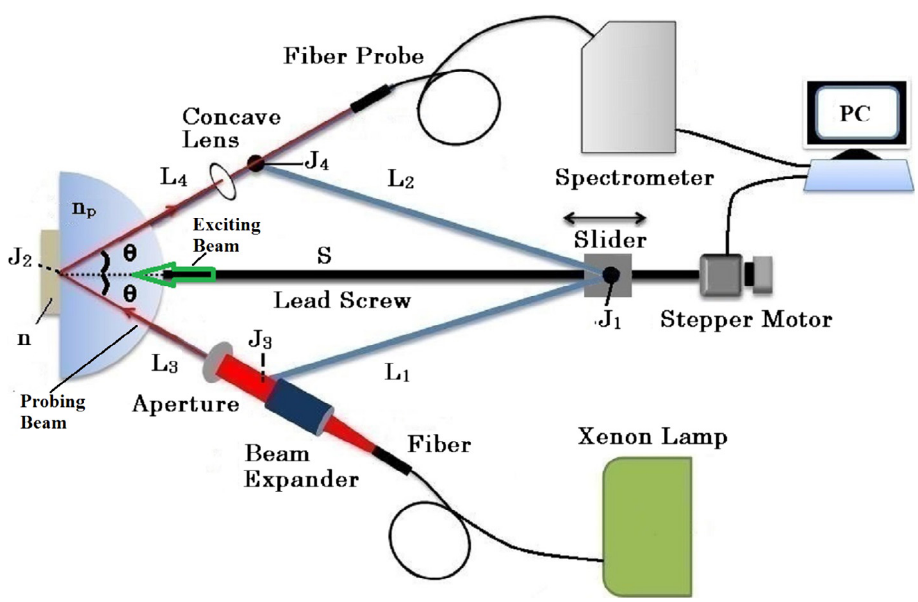


**Fig. 1.** Experimental apparatus.