

## Measuring Multiphoton Absorption Spectra with Thermal Lens Detection under A Crossed-Beam Configuration

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### Abstract

An experimental setup of excitation wavelength-tunable thermal lens detection under a crossed-beam configuration is constructed to measure multiphoton absorption spectra of molecules in a liquid solution. Dependence of the thermal lens signal intensity on light intensity for an aqueous solution of auramine O was nonlinear and showed one- and two-photon absorption. The thermal lens spectrum of the solution at 0.2 mJ/pulse of light intensity was measured for 410-600 nm.

### 1. Introduction

Molecules can absorb some photons simultaneously to be excited to a high energy state when the photon density becomes large. Even the visible and infrared light can excite relatively small molecules that have absorption wavelength in the UV region. The process is known as multiphoton absorption<sup>[1]</sup> and is frequently applied to two-photon excitation of fluorescence.

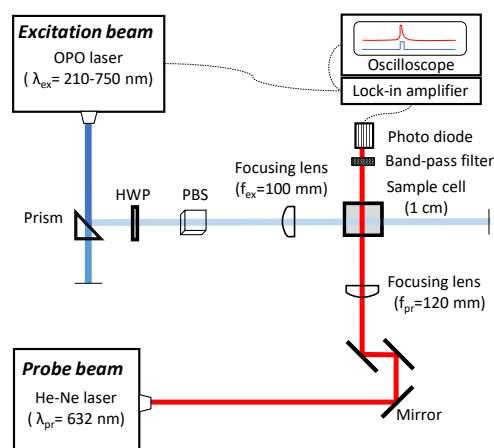
Thermal lens (TL) spectroscopy<sup>[2]</sup> is applied to analyze various materials using a change in a refractive index by thermal expansion after non-radiative relaxation of photoexcitation. Highly sensitive detection of non-fluorescent molecules in a liquid solution is one of the excellent applications of the method. Multiphoton excitation spectra of non- or less-fluorescent molecules are not widely investigated. Thus, an experimental setup of excitation-wavelength tunable thermal lens detection under a crossed-beam configuration is constructed to measure multiphoton absorption spectra of molecules in a liquid solution.

### 2. Experimental Methods

#### 2.1. Materials

Aqueous solutions of auramine O were selected as the sample. The solution concentrations were adjusted to  $1.0 \times 10^{-6}$  M for UV-Vis spectrophotometer and  $1.0 \times 10^{-4}$  M for TL spectrometer. The absorption peaks were 370 nm and 423 nm in the visible region.

#### 2.2. Experimental Setup



**Figure 1.** Experimental setup of the thermal lens spectroscopy under a crossed-beam configuration for measuring multiphoton absorption spectra.

HWP, half-wavelength-plate.

PBS, polarizing beam splitter.

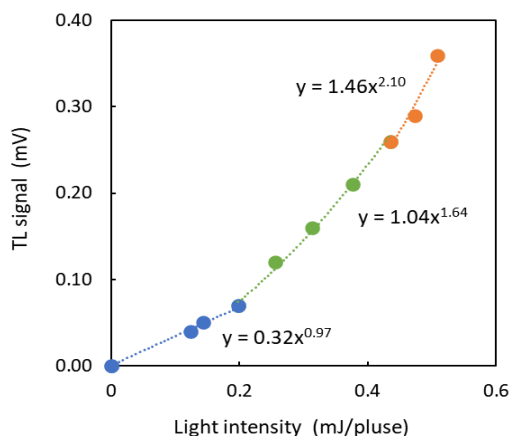
Figure 1 is a schematic illustration of the experimental setup of the thermal lens spectroscopy under a crossed-beam configuration. An output from a nanosecond-pulsed optical parametric oscillator (OPO. NT230-50-SH/SF-2H-3H, EXSPLA, repetition frequency, 50 Hz; pulse width, 5 ns) was used as an excitation beam, and the wavelength is tunable from 210 to 2006 nm.

The excitation beam was sequentially set a wavelength from 410 nm to 600 nm for measuring absorption spectra with the TL detection. The laser intensity was adjusted using a half-wavelength plate and a

polarizing beam splitter. One lens with a focal length of  $f_{\text{ex}}=100$  mm focused the excitation beam. The beam changed a refractive index to cause a TL effect. A He-Ne laser (LGK 7654-08, LASOS) was used as a probe beam of 632 nm wavelength, focused with another lens with a focal length of  $f_{\text{pr}}=120$  mm. The probe beam and the excitation beam crossed in a sample cell.

### 3. Results and Discussion

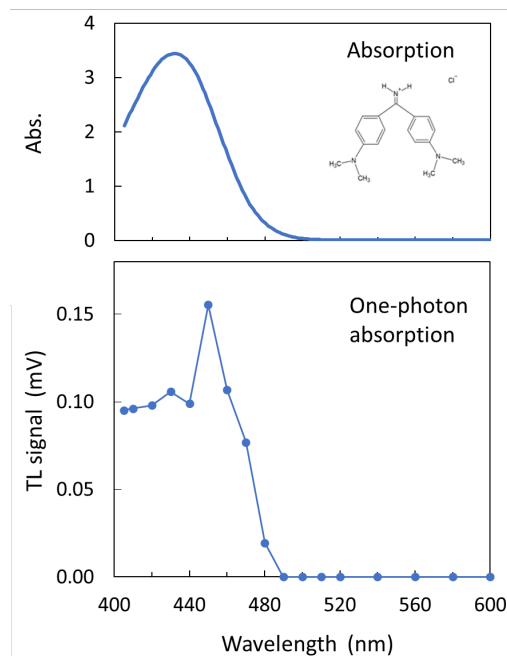
Figure 2 shows the dependence of the thermal lens signal intensity on light intensity (LI) for an aqueous solution of Auramine O at 432 nm wavelength. The graph shows a nonlinear behavior and seems to be divided into three regions. The low LI (0 - 0.2 mJ/pulse) region was caused by one-photon absorption, and the high LI (0.45 - 0.5 mJ/pulse) region was caused by two-photon absorption. The intermedial LI (0.2 - 0.45 mJ/pulse) region was caused by both one- and two-photon absorption. The LI of 0.2 mJ/pulse was selected to get a one-photon absorption spectrum.



**Figure 2.** Dependence of the thermal lens signal intensity on light intensity for an aqueous solution of auramine O.

Figure 3 shows the two types of absorption spectrum. The top one is the UV-Vis spectrum, and the bottom one is the TL spectrum at the LI of 0.2 mJ/pulse. The two spectra show a similar shape, and this TL spectroscopy obtained a one-photon absorption spectrum was measured with the TL spectroscopy constructed, and a two-photon

absorption signal was observed. However, measurement of the two-photon absorption spectrum suffered from the intensity instability of the OPO laser depending on the wavelength at the present stage.



**Figure 3.** Absorption spectra of aqueous solutions of auramine O. (Top) UV-Vis spectrum; (Bottom) thermal lens spectrum.

### 4. Conclusion

An experimental setup of excitation-wavelength tunable thermal lens detection under a crossed-beam configuration is constructed using a nanosecond-pulsed optical parametric oscillator as the excitation light source molecules in a liquid solution. Nonlinear behavior of excitation-power dependence of thermal lens signal intensity for an aqueous solution auramine O was observed. The thermal lens spectrum of the solution at 0.2 mJ/pulse of light intensity was measured.

### References

- [1] Joseph W. Perry, "Two-photon absorption: An overview of measurements and principles", *Adv. In optics and Photonics*, **2010**
- [2] A.Marcano, OK.Williams N.Melikechi, *Optics communications*, **2008**, 2595, 281

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