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Two-Photon Absorption of Electroluminescent Conducting Polymers

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Two-Photon Absorption of Electroluminescent Conducting Polymers

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We have investigated two-photon absorption(2PA) characteristics of highly efficient electroluminescent polymers, poly(phenylenevinylene) derivatives with an alkoxy and carbazole or oxadiazole groups as pendants: OxdEh-PPV and CzEh-PPV. We measured two-photon excitation spectra and compared them with one-photon excitation spectra. We found that two- and one-photon transitions satisfy different selection rules. The 2PA coefficients were obtained by measuring the photoluminescence spectra after two- and one-photon absorption. At 840nm, 2PA coefficients of OxdEh-PPV and CzEh-PPV are measured to be $1.0 \times 10^{-49} \text{ cm}^4 \cdot \text{s}$ and $4.4 \times 10^{-49} \text{ cm}^4 \cdot \text{s}$, respectively.

Key words: two-photon absorption, photoluminescence excitation, photoluminescence, PPV derivatives, oxadiazole, carbazole

INTRODUCTION

Electroluminescent polymers are attracting considerable attention due to possible applications as large-area light-emitting displays.¹⁻³⁾ There have been many efforts to improve the electroluminescence efficiency. In order to design the polymer with high efficiency, it is necessary to understand the luminescence mechanism. In this contribution, we have investigated two-photon absorption(2PA) characteristics of highly efficient electroluminescent polymers, poly(phenylenevinylene) derivatives with an alkoxy and carbazole or oxadiazole groups as pendants(Fig.1). To find the origin of the 2PA, we measured two-photon excitation(2PE) spectra and compared them with corresponding one-photon excitation(1PE) spectra. The 2PA coefficients were obtained by measuring the photoluminescence(PL) spectra after two- and one-photon absorption.

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EXPERIMENTAL RESULTS AND DISCUSSION

Two electroluminescent polymers were prepared for this experiment: OxdEh-PPV and CzEh-PPV (Fig. 1). OxdEh-PPV [poly[2-(4-[5-(4-*t*-butylphenyl)-1,3,4-oxadiazolyl] phenyl)-5-(2-ethylhexyloxy)-1,4-phenylene vinylene] and CzEh-PPV [poly[2-(*N*-carbazolyl)-5-(2-ethylhexyloxy)-1,4-phenylene vinylene]. These polymers were prepared by Gilch-Wheelwright method.⁴⁾ The number average molecular weights of these polymers were measured to be 12,500 (OxdEh-PPV) and 49,000 (CzEh-PPV) using GPC. The experiment in solution was performed with dilute solutions (5.0×10^{-4} M) of OxdEh-PPV and CzEh-PPV dissolved in 1,1,2,2-tetrachloroethane and tetrahydrofuran, respectively.

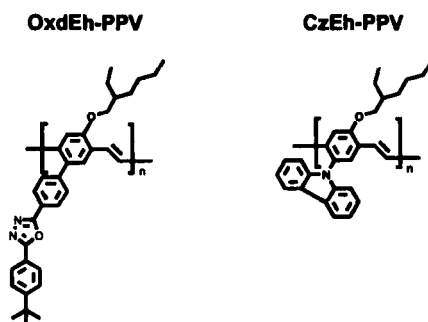


FIGURE 1. Chemical structures of PPV derivatives, OxdEh-PPV and CzEh-PPV.

To find the origin of the 2PA, we measured 2PE and 1PE spectra. The 2PE spectra were obtained with the optical parametric oscillator (OPO) pumped with the third harmonics of a nanosecond neodymium-doped yttrium-aluminum-garnet (Nd:YAG) laser and the 1PE spectra were acquired using a xenon lamp and a monochromator. The detection wavelength was fixed near the maximum photoluminescence position and the input power was fixed at 100 μ J. The excitation wavelengths were varied in the absorptive region. Data acquisition has been performed using a photomultiplier tube, a boxcar averager and a computer. To improve the signal-to-noise ratio, we averaged data over 2000 laser-shots. For the photoluminescence-excitation spectroscopy in the solution, the photoluminescence was collected in the perpendicular direction with respect to the direction of excitation beam. To obtain the 2PA coefficients, we measured the photoluminescence spectra after two- and one-photon absorption.⁵⁾ Recordings of the PL spectra were performed using a spectrophotometer. The excitation wavelengths were 840 nm and 420 nm for 2PA-PL and 1PA-PL, respectively. The PL spectra were obtained with the excitation energy of 6.3 mJ and 10 μ J for 2PA-PL and 1PA-PL, respectively.

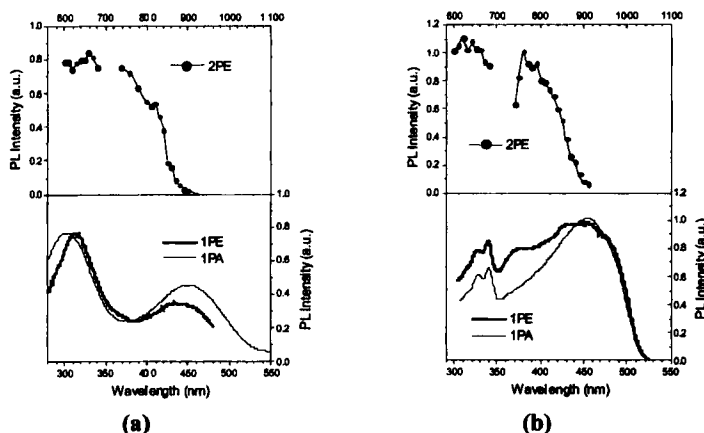


FIGURE 2. Two- and one-photon excitation spectra of OxdEH-PPV (a) and CzEH-PPV (b). Also, one-photon absorption spectra are shown.

Figure 2 shows 2PE spectra for OxdEH-PPV and CzEH-PPV. Two-photon excitation spectrum of OxdEH-PPV (Fig. 2-(a)) shows a plateau in the wavelength region between 600 nm and 820 nm. Above 820 nm, the 2PE signal decreases gradually until it disappears at 910 nm. Meanwhile, one-photon excitation spectrum has two broad bands at 443 nm (E1) and 313 nm (E2). Compared these bands with those of PPV and oxadiazole, the E1 band is thought to be due to photoluminescence by excitation of the phenylenevinylene unit in the main chain and the E2 band to the photoluminescence by excitation of the oxadiazole unit in the side chain. Also, the absorption spectrum has two bands in the wavelength region that are similar to those of excitation spectrum. We can understand these results with the selection rules. If we assign the E1 band of 1PE spectrum and the plateau of 2PE spectrum to the $1A_g \rightarrow 1B_u$ and the $1A_g \rightarrow nA_g$ transitions, respectively, then the 2PE spectrum shows that one-photon-allowed transition ($1A_g \rightarrow 1B_u$) disappears and two-photon-allowed transition ($1A_g \rightarrow nA_g$) appears.

Two-photon excitation spectrum for CzEH-PPV (Fig. 2-(b)) shows two bands. Meanwhile, one-photon excitation spectrum has three broad bands: E1, E2, and E3. The E1 band has two peaks at 430 nm and 456 nm and E2 band shows two peaks at 328 nm and 341 nm. Compared these bands with those of PPV and carbazole, the E1 band is thought to be due to photoluminescences by excitation of the phenylenevinylene unit in the main chain and the E2 band to photoluminescence by excitation of the carbazole unit in the side chain. A new band (E3) appears around 372 nm and its origin is not yet clearly understood. Similarly with 2PE of OxdEH-PPV, the 2PE of CzEH-PPV shows the two-photon transition ($1A_g \rightarrow nA_g$) that is shifted to the blue side when compared with the one-photon transition ($1A_g \rightarrow 1B_u$).

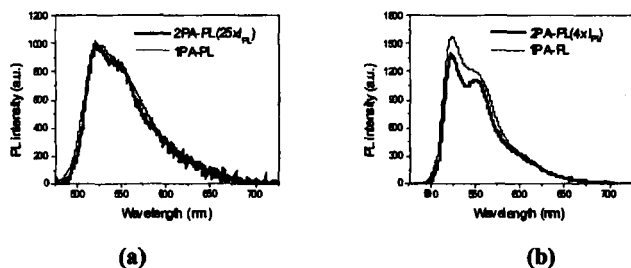


FIGURE 3. Photoluminescence spectra after two- and one-photon absorption: (a) OxdEh-PPV and (b) CzEh-PPV.

Shown in Fig. 3 are the photoluminescence spectra after two- and one-photon absorption. Both OxdEh-PPV and CzEh-PPV exhibit the 2PA-PL spectra that are similar to 1PA-PL spectra. The photoluminescence spectra of the OxdEh-PPV and CzEh-PPV show the spectroscopic features that can be resolved to two peaks. By comparing these spectra with the PL spectra of rhodamine 6G, we obtained the 2PA coefficients of $1.0 \times 10^{-49} \text{ cm}^4/\text{s}$ and $4.4 \times 10^{-49} \text{ cm}^4/\text{s}$ for OxdEh-PPV and CzEh-PPV, respectively.

In conclusion, it was found that the two- and one-photon transitions satisfy the different selection rules. Both OxdEh-PPV and CzEh-PPV showed the two-photon transition that was shifted to the blue side when compared with the one-photon transition. At 840 nm, 2PA coefficients of OxdEh-PPV and CzEh-PPV are measured to be $1.0 \times 10^{-49} \text{ cm}^4/\text{s}$ and $4.4 \times 10^{-49} \text{ cm}^4/\text{s}$, respectively.

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