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## The impact of the pi-electron conjugation length on the three-photon absorption cross section of fluorene derivatives

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# The impact of the $\pi$ -electron conjugation length on the three-photon absorption cross section of fluorene derivatives

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The three-photon absorption cross sections of three different fluorene derivatives, with extended  $\pi$ -electron conjugation lengths was experimentally measured and compared with shorter  $\pi$ -electron conjugation length analogs. The effect of the conjugation length on the three-photon absorption cross section  $\sigma'_3$  of this family of molecules has been elucidated. It is demonstrated that  $\sigma'_3$  of the asymmetric compound  $D$ - $\pi$ - $\pi$ - $\pi$ - $A$  is 6.6 times larger than its shorter configuration  $D$ - $\pi$ - $A$ , while for the symmetric compounds  $D$ - $\pi$ - $\pi$ - $\pi$ - $D$  and  $A$ - $\pi$ - $\pi$ - $\pi$ - $A$  a two-fold enhancement was found relative to their shorter conjugation length analogs. Measurements of the three-photon excitation of these compounds in THF solution ( $10^{-3}M$ ) were accomplished with a tunable optical pulse generation pumped by a 25 ps Nd-YAG laser. © 2006 American Institute of Physics.  
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## I. INTRODUCTION

The three-photon absorption (3PA) process has received considerable attention from the scientific community in the last decade because of its potential applications in photonics and biomedical fields.<sup>1-4</sup> One of the most important objectives in this field, at present, is to synthesize and characterize new molecules with high 3PA cross section.<sup>5-11</sup> In organic molecules, 3PA typically occurs at long wavelengths in the near-infrared region. Therefore, minimization of scattered light losses is expected,<sup>12</sup> and a reduction of undesirable linear absorption in living organisms that present semitransparent windows at approximately 1100 and 1300 nm (Ref. 13) is anticipated. Consequently, the radiation penetration depth through tissue can be maximized, thus facilitating tumor imaging and photoannihilation in the absence of complicated and risky surgery.

Although, 3PA seems to be a good candidate for biomedical applications, molecules with higher  $\sigma'_3$  are still in great need. However, at present there is a lack of systematic experimental studies on the structure-property relationship for 3PA that make possible molecular engineering to optimize the three-photon absorption cross section of organic materials.

During the last three years, we have been engaged in the methodical study of a series of highly conjugated fluorene derivatives with different structural motifs:  $D$ - $\pi$ - $A$ ,<sup>14</sup>  $D$ - $\pi$ -

$D$ ,<sup>11</sup> and  $A$ - $\pi$ - $A$ ,<sup>15</sup> where  $D$  and  $A$  represent electron-donor ( $N,N$ -diphenylamino) and electron-acceptor (benzothiazole) groups. So far, we have demonstrated that symmetric intramolecular charge transfer favors three-photon absorption in fluorene derivatives. The most effective structure turned out to be an  $A$ - $\pi$ - $A$  compound.<sup>15</sup> The high functionality of the fluorene aromatic ring provides the ability to introduce particular functionality through molecular engineering at positions 2 and 7.<sup>16-18</sup> As a result, using the fluorene core, one has the ability to generate different chemical archetypes by affecting the electron accepting and donating properties of the system as well as the possibility of extending the  $\pi$ -electron conjugation length.

In this paper we present the systematic study of the effect the  $\pi$ -electron conjugation length has on the three-photon absorption cross section of this family of fluorene derivatives. The  $\pi$ -electron conjugation length of each compound was extended by inserting two phenylene-vinylene groups, one on each side of the fluorenyl core.<sup>18</sup> The new compounds, 4-(( $E$ )-2-(7-(4-(benzo[d]thiazol-2-yl)styryl)-9,9-didecyl-9H-fluorene-2-yl)vinyl)- $N,N$ -dibutylaniline (( $D$ - $\pi$ - $\pi$ - $\pi$ - $A$ ), 7-diyl)bis(ethene-2,1-diyl)bis(4,1-phenylene))dibenzo[d]thiazole ( $D$ - $\pi$ - $\pi$ - $\pi$ - $D$ ), and 2,2'-(4,4'-(1 $E$ ,1' $E$ )-2,2'-(9,9-didecyl-9H-fluorene-2,7-diyl)bis(ethene-2,1-diyl)bis(4,1-phenylene))dibenzo[d]thiazole ( $A$ - $\pi$ - $\pi$ - $\pi$ - $A$ ) are characterized by a longer distance over which the electric charge can be transferred between donor and acceptor groups via conjugated bridge or, from the ends of the molecule to the conjugated core and vice versa. The increase in the

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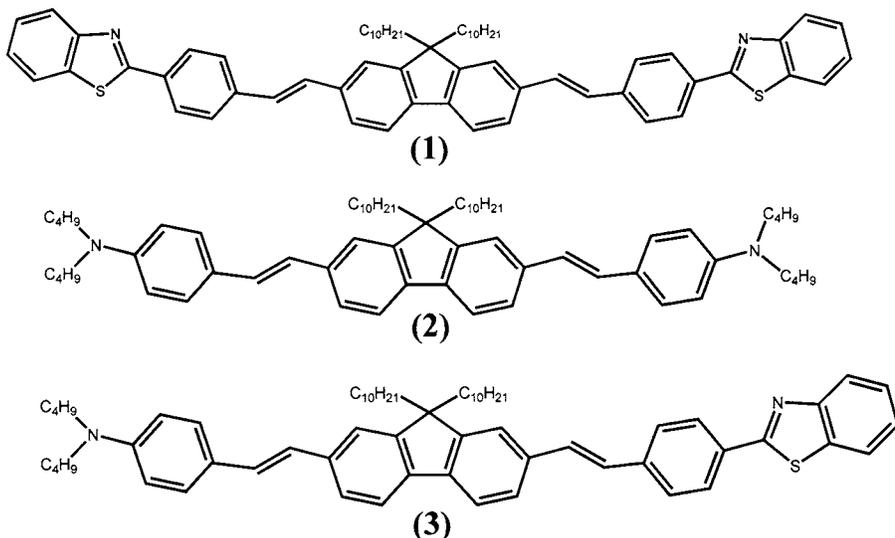


FIG. 1. Molecular structure of  $A$ - $\pi$ - $\pi$ - $\pi$ - $A$  (1),  $D$ - $\pi$ - $\pi$ - $\pi$ - $D$  (2), and  $D$ - $\pi$ - $\pi$ - $\pi$ - $A$  (3) compounds.

$\pi$ -electron conjugation length leads to a stronger  $\pi$ -electron delocalization. Therefore, an enhancement of the imaginary part of the fifth-order hyperpolarizability  $\chi_I^{(5)}(-\omega; \omega, -\omega, \omega, -\omega, \omega)$ , i.e., the three-photon absorption cross section, was anticipated.

## II. EXPERIMENTAL SECTION

The molecular structures of the three molecules studied here are shown in Fig. 1. The first compound,  $A$ - $\pi$ - $\pi$ - $\pi$ - $A$  (1), is symmetrical, and consists of a fluorenyl core, two phenylene-vinylene groups attached to the 2 and 7 positions, and one acceptor group (benzothiazole) at each of the two extremities. The second compound,  $D$ - $\pi$ - $\pi$ - $\pi$ - $D$  (2), is also symmetrical, but the end groups are electron donors ( $\text{NC}_8\text{H}_{18}$ ). The last compound,  $D$ - $\pi$ - $\pi$ - $\pi$ - $A$  (3), is an unsymmetrical molecule with a benzothiazole group on one side and a  $\text{NC}_8\text{H}_{18}$  group on the other. The detailed synthesis of the symmetric compound  $A$ - $\pi$ - $\pi$ - $\pi$ - $A$  (1) is described in Ref. 18. The preparation method of (2) and (3) will be published elsewhere.

The absorption spectra of the organic dyes were measured using an Agilent 8453 diode array UV-vis spectrometer (cell effects and solvent contributions were properly subtracted). To carry out these measurements, a 1-mm-path quartz cell filled with a THF solution, at a concentration of  $10^{-5}M$  on the targeted compound, was employed. The linear emission spectral measurements were carried out using a JY-TRIAX 550 spectrophotometer exciting at 420 nm. The experimental setup for multiphoton absorption-induced fluorescence is presented in Fig. 2.

All multiphoton-induced fluorescence and degenerate three-photon absorption measurements were performed using an EKSPLA 401 optical pulse generation (OPG) system pumped with a mode-locked, 25 ps EKSPLA PL-2143B laser [full width at half maximum (FWHM)], working at 10 Hz repetition rate. After spatial filtering [lenses L1, L2, and the pinhole (PH)], the laser beam was directed to the sample (by mirrors M1 and M2) and, focused inside

the cell using lens L3. The fluorescence light from the organic dyes was collected with lens L4 placed perpendicular to the cell, and then coupled into the JY-TRIAX 550 spectrometer.

Using the well known open-aperture Z-scan technique,<sup>19</sup> we measured the three-photon absorption coefficient of each compound at different wavelengths, tuning the excitation wavelength within the  $S_0 \rightarrow S_1$  band, over a range of 100 nm. We obtained a peak 3PA coefficient for the  $A$ - $\pi$ - $\pi$ - $\pi$ - $A$ ,  $D$ - $\pi$ - $\pi$ - $\pi$ - $D$ , and  $D$ - $\pi$ - $\pi$ - $\pi$ - $A$  molecules pumping at 1230, 1280, and 1260 nm, respectively. For all Z-scan measurements a 1-mm-path-length cell was used, filled with the corresponding organic dye in a THF solution at a concentration of  $10^{-3}M$ . Typical irradiances above  $45 \text{ GW/cm}^2$  were employed for 3PA measurements. Using the setup presented in Fig. 2, we demonstrated three-photon absorption-induced fluorescence at the excitation wavelengths mentioned above for each compound. In order to confirm the three-photon absorption process, we measured the transmitted intensity versus incident intensity holding the sample at the focal point of the focusing lens L3. The laser beam was separated into

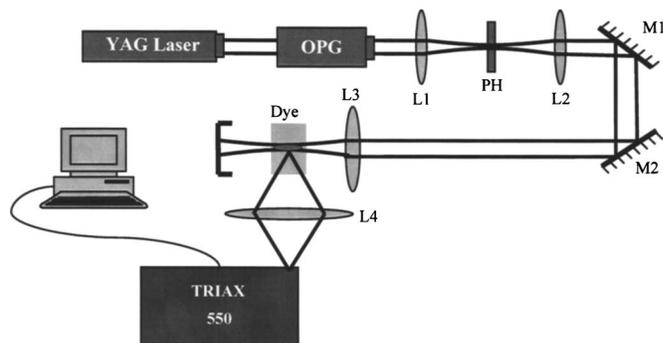


FIG. 2. Experimental setup for multiphoton induced fluorescence. The picosecond laser is pumping an EKSPLA 401 OPG system which provide wavelength from  $420 \mu\text{m}$  to  $2.3 \mu\text{m}$ . The fluorescence light is collected by lens L4 and coupled into a JY-TRIAX 550 spectrometer. 1-cm-path quartz cell, filled with the dyes solution in THF ( $10^{-3}M$ ) was used for fluorescence measurements.

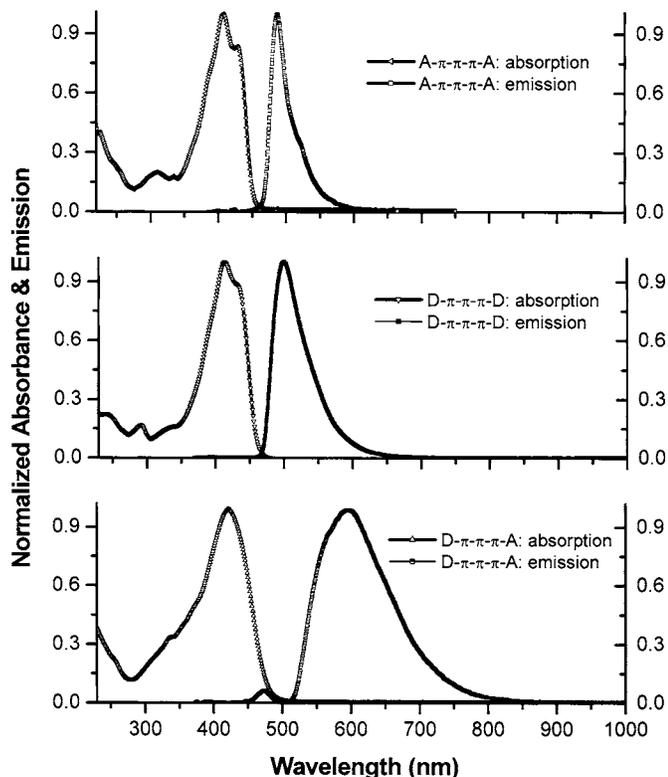


FIG. 3. Linear absorption (triangles) and emission (squares) spectra. The linear absorption spectra were measured in THF at a concentration of  $10^{-5}M$  using an Agilent 8453 diode array spectrometer. Linear emission was recorded with a high performance JY-TRIAx 550 spectrometer.

two beams (using a beam splitter), one weak beam for the reference and a strong focused beam for the signal. Because near-infrared (NIR) radiation was employed, germanium photodiode detectors were used. The incident intensity was gradually increased from  $5 \text{ GW/cm}^2$  up to  $90 \text{ GW/cm}^2$  using neutral density filters. The excitation wavelength was tuned from 1230 to 1260 to 1280 nm, corresponding to the spectral wavelengths for the peak 3PA cross section of each compound, as determined by Z-scan measurements.

### III. RESULTS AND DISCUSSIONS

Figure 3 displays the linear absorption and emission spectra of each fluorene derivative studied. Each compound shows strong linear absorption in the spectral range of 300–500 nm, with an absorption maximum at 410, 415, and 425 nm, for  $A-\pi-\pi-\pi-A$ ,  $D-\pi-\pi-\pi-D$ , and  $D-\pi-\pi-\pi-A$  derivatives, respectively. These wavelengths correspond to the transition between the ground state and first excited state  $S_0 \rightarrow S_1$ .<sup>18</sup> Also, it was observed that above 500 nm there was

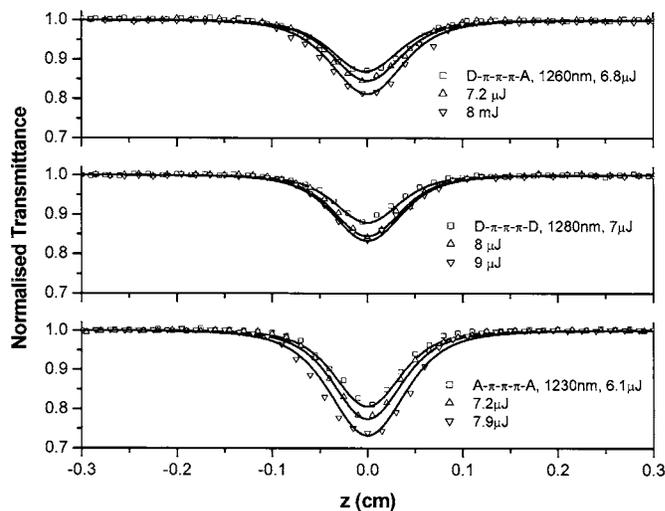


FIG. 4. Open-aperture Z-scan curves of  $D-\pi-\pi-\pi-A$ ,  $D-\pi-\pi-\pi-D$ , and  $A-\pi-\pi-\pi-A$  in THF ( $10^{-3}M$ ), taken at three different energies for each compound. Solid curves are the best theoretical fittings using Gaussian beam equations for 3PA. Measurements were done using 1-mm-path cell and the same tunable EKSPLA 401 OPG system pumped by the third harmonic of a mode-locked, 25 ps full width at half maximum (FWHM), Nd-YAG (yttrium aluminum garnet) laser (EKSPLA PL-2140B), operating at a 10 Hz repetition rate. The beam waist at the focal plane was ca. 14.25, 14.55, and  $13.75 \mu\text{m}$  ( $\text{HW}1/e^2M$ ) at  $\lambda_p=1260$ , 1280, and 1230 nm, respectively.

no linear absorption for any of the three molecules. The one-photon fluorescence spectrum of each compound is shown in Fig. 3 by square symbols. The fluorescence emission band shows a peak wavelength at 489, 500, and 596 nm, for the  $A-\pi-\pi-\pi-A$ ,  $D-\pi-\pi-\pi-D$ , and  $D-\pi-\pi-\pi-A$  derivatives, respectively. The symmetrical compounds exhibited Stoke's shift smaller than that of the unsymmetrical compound. This result was expected because of the small change in the molecular dipole moment upon electronic excitation for symmetrical molecules.<sup>18</sup>

Figure 4 shows both the experimental Z-scan curves given by points and the theoretical fitting curves represented by solid lines. To fit the experimental curves, we employed the equations for a three-photon absorption process considering temporal and spatial Gaussian profiles, derived by Co-hanoschi and Hernandez.<sup>20</sup> For all three compounds, the 3PA coefficient was measured employing three different input energies. The relative error of the absorption coefficient was approximately 14%. The corresponding 3PA coefficients were  $\alpha_3=(5.5\pm 0.8)\times 10^{-21} \text{ cm}^3 \text{ W}^{-2}$  for  $D-\pi-\pi-\pi-A$  pumping at 1260 nm,  $\alpha_3=(3.9\pm 0.5)\times 10^{-21} \text{ cm}^3 \text{ W}^{-2}$  for  $D-\pi-\pi-\pi-D$  pumping at 1280 nm, and  $\alpha_3=(1.18\pm 0.16)\times 10^{-20} \text{ cm}^3 \text{ W}^{-2}$  for the  $A-\pi-\pi-\pi-A$  molecule pumping at

TABLE I. Three-photon absorption cross sections and the corresponding excitation wavelengths for all compounds.

Compound	$D-\pi-A$	$D-\pi-D$	$A-\pi-A$	$D-\pi-\pi-A$	$D-\pi-\pi-D$	$A-\pi-\pi-A$
Excitation wavelength (nm)	1200	1110	1095	1260	1280	1230
$\sigma_3^a$ ( $\text{cm}^6 \text{ s}^2/\text{photon}^2$ )	37 <sup>a</sup>	82 <sup>a</sup>	335 <sup>b</sup>	245	145	511

<sup>a</sup>Measured values given in Ref. 9.

<sup>b</sup>Values reported in Ref. 11.

1230 nm. The three-photon absorption coefficient can be correlated to the three-photon absorption cross section by

$$\sigma'_3 = \frac{\alpha_3}{N_A d_0 10^{-3}} \left( \frac{hc}{\lambda} \right)^2, \quad (1)$$

where  $N_A$  is Avogadro's number ( $N_A$ ),  $d_0$  (mol/l) is the solution concentration, and  $hc/\lambda$  is the energy of the incident photons.<sup>21</sup> Using equation (1), the following values were obtained for the three-photon absorption cross section:  $511 \times 10^{-78}$ ,  $145 \times 10^{-78}$  and  $245 \times 10^{-78}$  cm<sup>6</sup> s<sup>2</sup>/photon<sup>2</sup> for  $A$ - $\pi$ - $\pi$ - $\pi$ - $A$ ,  $D$ - $\pi$ - $\pi$ - $\pi$ - $D$ , and  $D$ - $\pi$ - $\pi$ - $\pi$ - $A$ , respectively. These values indicate that  $\sigma'_3$  is enhanced by a factor of 1.6, 1.8, and 6.6 with respect to the corresponding shorter  $\pi$ -electron conjugation length molecules  $A$ - $\pi$ - $A$ ,  $D$ - $\pi$ - $D$ , and  $D$ - $\pi$ - $A$ , respectively (see Table I). In Table I, the 3PA cross sections are presented corresponding to both, longer and shorter  $\pi$ -electron conjugation length analogs and the corresponding excitation wavelengths.

The increase of  $\pi$ -electron conjugated bridge length by inserting phenylene-vinylene groups leads to an extension of the distance over which the electric charge can be transferred. Therefore, an increase of the  $\pi$ -electron delocalization in the first excited state  $S_1$  is induced. This electronic delocalization will enhance the  $S_0 \rightarrow S_1$  transition dipole moment, which makes an important contribution to the 3PA cross section.<sup>22</sup>

The second method utilized to measure the three-photon absorption cross section involved intensity-dependent transmittance measurements, as described in the Experimental section. Neglecting the linear absorption at the pump wavelength, the beam attenuation along the optical propagation path  $z$  due to three-photon absorption is given by the following equation:

$$\frac{dI(z)}{dz} = -\alpha_3 I(z)^3. \quad (2)$$

Here  $\alpha_3$  is the 3PA coefficient of the sample at the pump wavelength and  $I(z)$  is the irradiance that depends on the propagation distance  $z$ . The straightforward solution of Eq. (2) is

$$I(z) = \frac{I_0}{\sqrt{1 + 2\alpha_3 z I_0^2}}, \quad (3)$$

where  $I_0$  is the input irradiance of the excitation beam. Equation (3) can be numerically solved for  $\alpha_3$ . Thus, the cross section can be determined. Figure 5 shows the transmitted intensity versus input intensity curves for each of the three fluorene derivatives. The solid lines represent the theoretical fittings with the best-fit parameter  $\alpha_3 = 2.5 \times 10^{-21}$  cm<sup>3</sup> W<sup>-2</sup>,  $\alpha_3 = 1.32 \times 10^{-21}$  cm<sup>3</sup> W<sup>-2</sup>, and  $\alpha_3 = 1.18 \times 10^{-21}$  cm<sup>3</sup> W<sup>-2</sup> for  $A$ - $\pi$ - $\pi$ - $\pi$ - $A$ ,  $D$ - $\pi$ - $\pi$ - $\pi$ - $D$ , and  $D$ - $\pi$ - $\pi$ - $\pi$ - $A$ , respectively. These values are of the same order of magnitude as the values obtained using  $Z$  scan. However, they are three to four times smaller. The discrepancy between the two methods is given by the fact that the theoretical fitting for the second method assumes a square temporal profile instead of the actual Gaussian distribution.

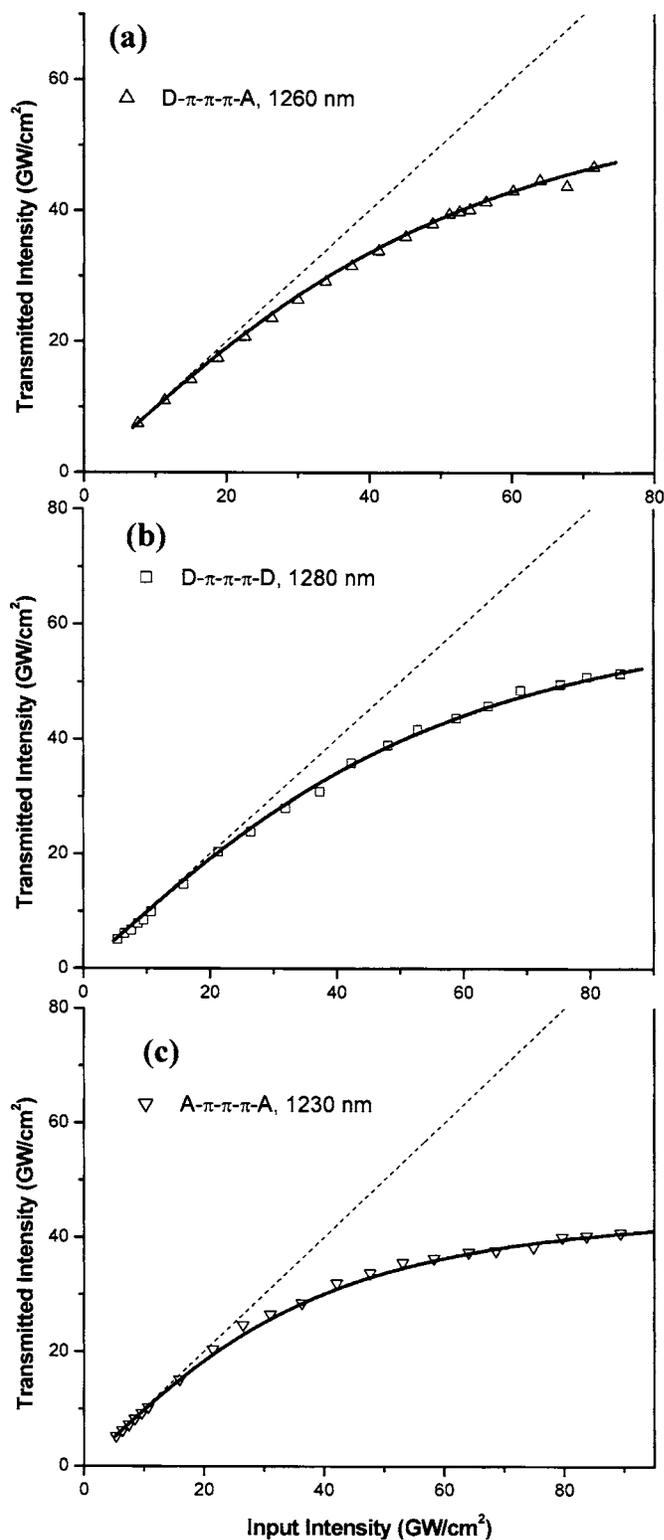


FIG. 5. Transmitted intensity vs input intensity curves of  $D$ - $\pi$ - $\pi$ - $\pi$ - $A$  (a),  $D$ - $\pi$ - $\pi$ - $\pi$ - $D$  (b), and  $A$ - $\pi$ - $\pi$ - $\pi$ - $A$  (c). The solid lines represent the theoretical fittings using Eq. (3). The best-fit parameters were  $\alpha_3 = 1.18 \times 10^{-21}$  cm<sup>3</sup> W<sup>-2</sup> (a),  $\alpha_3 = 1.32 \times 10^{-21}$  cm<sup>3</sup> W<sup>-2</sup> (b), and  $\alpha_3 = 2.5 \times 10^{-21}$  cm<sup>3</sup> W<sup>-2</sup> (c). The excitation wavelengths in each case were 1260 nm (a), 1280 nm (b), and 1230 nm (c). The dashed lines are the fitted curves with  $\alpha_3 = 0$ .

The three-photon absorption-induced fluorescence was measured for the three fluorene derivatives using the setup illustrated in Fig. 2. Figure 6 shows the 3PA-induced fluo-

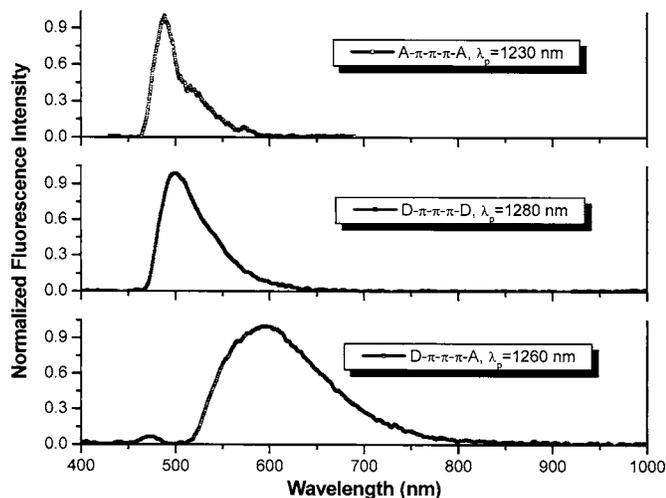


FIG. 6. Three-photon absorption-induced fluorescence spectra of  $A-\pi-\pi-\pi-A$  (open squares),  $D-\pi-\pi-\pi-D$  (filled squares), and  $D-\pi-\pi-\pi-A$  (semiopen squares) pumping at 1230, 1280, and 1260 nm, respectively.

rescence spectra of  $A-\pi-\pi-\pi-A$ ,  $D-\pi-\pi-\pi-D$ , and  $D-\pi-\pi-\pi-A$ , pumping at excitation wavelengths of 1230, 1280, and 1260 nm, respectively. The results indicate the maximum emission intensity at 489, 502, and 596 nm, corresponding to  $A-\pi-\pi-\pi-A$ ,  $D-\pi-\pi-\pi-D$ , and  $D-\pi-\pi-\pi-A$ , in that order. Virtually identical spectra were obtained for all three compounds (see Fig. 3) using single-photon excitation, relative to three-photon excitation. This indicates that the final excited state is independent of the excitation wavelength, and the excitation takes place between the ground state  $S_0$  and first excited state  $S_1$ . Because, there is no intermediate real state between  $S_0$  and  $S_1$ , and these compounds do not undergo linear absorption at excitation wavelengths above 500 nm, therefore, the measured nonlinear process can be attributed to pure three-photon absorption.

#### IV. CONCLUSIONS

In summary, we have demonstrated an enhancement of the three-photon absorption cross section of both symmetric and asymmetric fluorene derivatives by extending the  $\pi$ -electron conjugation length of this family of molecules. The asymmetric compound  $D-\pi-\pi-\pi-A$  exhibited the largest enhancement relative to the shorter  $\pi$ -electron conjugation length fluorene derivative  $D-\pi-A$ . The 3PA cross sections were measured using two different techniques,  $Z$ -scan and intensity-dependent transmittance measurements. Also, the three-photon absorption-induced fluorescence was demonstrated for all three fluorene derivatives. In terms of molecu-

lar design, our results suggest that the 3PA cross section of this family of molecules can be enhanced by increasing the distance over which the electric charge can be transferred. It is expected that the biophotonics field will benefit from new fluorophores that exhibit extremely high 3PA.

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