Electronic Supplementary Information (ESI)

Second Harmonic Generation in Laterally Azo-Bridged H-Shaped Ferroelectric Dimesogens†

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1. Estimation of the hyperpolarizability of DR-1 chromophore at 1.6 μm

It was reported that the β value of DR-1 chromophore at 1.9 μm is $\beta(1.9) = 49 \times 10^{-30}$ esu. According to the two-level model, the β value of DR-1 chromophore at 1.6 μm is estimated by the following equation:

$$
\frac{\beta(\lambda')}{\beta(\lambda')} = \frac{1 - \left(\frac{2\lambda_0}{\lambda'}\right)^2}{1 - \left(\frac{2\lambda}{\lambda'}\right)^2} \frac{1 - \left(\frac{\lambda_0}{\lambda'}\right)^2}{1 - \left(\frac{\lambda}{\lambda'}\right)^2},
$$

(a)

where $\lambda_0$ is the resonator wavelength. In this specific case, $\lambda' = 1.6 \mu m$, $\lambda = 1.9 \mu m$, and $\lambda_0 = 0.53 \mu m$ (the maximum absorption wavelength of compound 2). The β value at 1.6 μm is obtained, $\beta(1.6) = 62 \times 10^{-30}$ esu.

2. The origin of Eq. (4)

The equation (4) is obtained by the transformation of a third rank tensor when the reference frame is rotated by an angle $\theta$ (i.e., the tilt angle) about the y-axis (see the
transformation in Fig. S1). In general the transformation of the third rank tensor is expressed as

$$\beta^{(d)}_{i,j,k} = \sum_{i,j,k} A_{ik} A_{jk} A_{ki} \beta^{(d)}_{i,j,k}.$$  \hfill (b)

where all indices go from 1 to 3.

In the particular case the rotation matrix is

$$A = \begin{pmatrix} \cos \theta & 0 & -\sin \theta \\ 0 & 1 & 0 \\ \sin \theta & 0 & \cos \theta \end{pmatrix}.$$  \hfill (c)

Take $\beta^{(d)}_{yzz} = \beta^{(d)}_{233}$ as an example. $\beta^{(d)}_{233}$ is expressed as

$$\beta^{(d)}_{233} = \sum_{i,j,k} A_{ij} A_{jk} A_{ki} \beta^{(d)}_{i,j,k} = A_{y} A_{x} A_{z} \beta_{221}^{(d)} + A_{y} A_{z} A_{x} \beta_{211}^{(d)} + A_{z} A_{x} A_{y} \beta_{213}^{(d)} + A_{z} A_{y} A_{x} \beta_{233}^{(d)}$$

$$= A_{y} A_{x} A_{z} \beta_{221}^{(d)} + A_{z} A_{x} A_{y} \beta_{211}^{(d)} + \sin^2 \theta \beta_{xz}^{(d)} + \cos^2 \theta \beta_{zz}^{(d)}.$$  \hfill (d)

Then the $d_{23}$ (i.e., $d_{233}$ in three-indices notation) coefficient is

$$d_{23} = N f^3 \beta_{xzz}^{(d)} = N f^3 (\beta_{xzz}^{(d)} \sin^2 \theta + \beta_{zzz}^{(d)} \cos^2 \theta).$$  \hfill (e)

The other three $d$ tensor components, $d_{22}$, $d_{21}$, and $d_{14}$ are obtained as shown in eq. (4) by following the same operation as shown above.