

Modeling of the second harmonic generation in SiO₂ sol–gel films doped with nanoscopic DR1 molecules as function of the poling time

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Available online 3 May 2006

Abstract

A physical description for the second harmonic generation (SHG) intensity as function of corona poling time in SiO₂:DR1 sol–gel films is shown. This model describes the poling time dependence of the SHG based on a damped oscillator description of the nanomolecules of DR1. Instead of an usual biexponential or polynomial model with meaningless parameters, all the parameters from our model have a physical interpretation. The model results have been experimentally verified in several SiO₂:DR1 sol–gel films with different side-chain doping concentrations.

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PACS: 42.70.Jk; 78.20.Bh; 78.40.Me

Keywords: Sol–gel; Chromophores; Corona poling; Second harmonic generation

1. Introduction

Interesting macroscopic optical properties can be obtained by controlling the orientation of nanoscopic molecules inside of materials. Functionalized polymers have been extensively studied to incorporate non-linear optical (NLO) chromophores into macromolecular systems. For an optimization of the second-order non-linear optical response in an assembly of NLO active molecules [1], a high degree of non-centrosymmetric order of the dipoles is needed. It can be attained by either electric field [2] or single-point Corona poling technique field [3]. In the last case an important goal is to get the alignment optimization of the dopant dipolar molecules under the influence of a

d.c. external applied field. In order to do that, some studies have been performed to optimize the orientation by taking into account the relationship between molecular and macroscopic properties of orientationally ordered materials [4,5], the intermolecular electrostatic [6–8] and the chromophore–matrix interactions [9,10]. Some models have been developed to describe the time dependence of this alignment [5,11] or even the time and temperature dependence of its relaxation [12].

In this work, we modeled the orientation dependence with respect to the poling time considering the equation of a damped oscillator. The model considers implicitly the chromophore–matrix interactions into the damping constant. We use this model to fit the behavior of the second harmonic generation intensity as function of the poling time in sol–gel films with side-chain DR1 (1.5 nm length) doping. The model describes the increment in second harmonic generation intensity as corona poling time increases until getting a saturation value for large poling times.

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In Section 2 we develop the model and describe the considerations taken into account for applying it to the films. Section 3 contains the material preparation and the experimental methods for poling the samples and the measurements of the SHG intensity. The results are presented and discussed in Section 4. Finally, we give our conclusions in Section 5.

2. Theory

2.1. Basics

The SHG intensity of a system of organic molecules is strongly dependent of the molecular geometrical arrangement. SHG exists only if the geometrical arrangement of the organic molecules system gives place to a macroscopic non-centrosymmetry. This situation is achieved in polymer-like materials by orienting their large permanent dipole moment organic molecules by means of a strong electrostatic field, like Corona field.

The non-centrosymmetric condition gives place to the second order molecular hyperpolarizability β_{ijk} constant and to the molecular system second order susceptibility function $\chi_{LJK}^{(2)}(t)$ as the most important terms for the molecular system SHG.

The microscopic polarization for only one molecule can be written as

$$p_i(t) = \mu_i^0 + \alpha_{ij}E_j(t) + \beta_{ijk}E_j(t)E_k(t) + \gamma_{ijkl}E_j(t)E_k(t)E_l(t) + \dots, \quad (1)$$

with p_i the microscopic polarization along the i axis of the molecule, $E_{j,k,l,\dots}$ the electric field acting on the molecule along a j,k,l,\dots molecular axis, which in general have a temporal dependence, μ_i^0 is the permanent dipole moment along the i molecular axis, α_{ij} is the linear polarizability constant of the molecule, β_{ijk} is the molecular second order hyperpolarizability constant, γ_{ijkl} is the molecular third order hyperpolarizability constant, and t is the time variable.

The macroscopic polarization for a molecular system can be written as

$$P_I(t) = P_I^{(0)}(t) + \chi_{IJ}^{(1)}(t)E_J(t) + \chi_{LJK}^{(2)}(t)E_J(t)E_K(t) + \chi_{LJKL}^{(3)}(t)E_J(t)E_K(t)E_L(t) + \dots, \quad (2)$$

with $P_I(t)$ the temporal-depending macroscopic polarization of the molecular system along the I axis, $P_I^{(0)}(t)$ the system spontaneous polarization, $\chi_{IJ}^{(1)}(t)$ the system first order susceptibility function, $\chi_{LJK}^{(2)}(t)$ the system second order susceptibility function, $\chi_{LJKL}^{(3)}(t)$ the system third order susceptibility function, and $E_{J,K,L,\dots}(t)$ the electric field acting on the molecular system along a J,K,L,\dots macroscopic reference system axis.

The existing relation between the microscopic and macroscopic reference systems is given by Wu [5] and is illustrated in Fig. 1. The microscopic axes are identified by

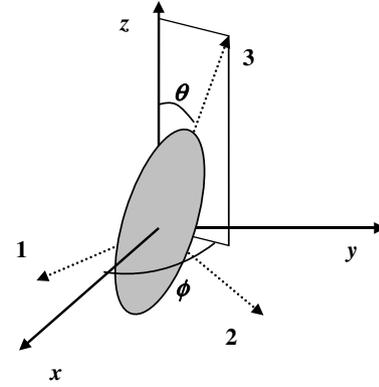


Fig. 1. Coordinate systems. Dashed lines indicates the molecular principal axes (1, 2, 3). Solid lines indicates the macroscopic laboratory coordinate axes (x, y, z).

numbers and the macroscopic axes are identified by letters, θ is the director angle between the z axis of both systems and ϕ is the azimuthal angle. The corona field is applied along the z axis. The macroscopic system has azimuthal symmetry.

The macroscopic orientation of a rod-like molecule with a large permanent dipole moment along its main principal axis (μ_3^0) under the influence of an external electrostatic field applied along the z axis (E_z^0) is well described by the θ angle, and is ϕ angle independent. Actually, θ angle is a poling-time dependent function, which at the beginning has an arbitrary initial value, and if there is not more fields than E_z^0 , θ angle should tend to zero as poling time increases. The polar angle would tend to other value as poling time increases if there were more fields than the E_z^0 one.

As a matter of fact, the spontaneous polarization and the susceptibilities in Eq. (2) are poling time dependents because the polar angle of each molecule of the system is poling time dependent. The susceptibilities have, really, the geometrical information of the molecular system. In consequence, to make an adequate description of the SHG intensity emerging from a polymer-like material as function of poling time, it is required to calculate its second order susceptibility function.

Since rod-like molecules usually have their most important second order hyperpolarizability constant term along its main principal axis (β_{333}), their second order polarizabilities can be written as

$$p_3^{(2)}(t) = \beta_{333}E_J(t) \cos(3, J)(t)E_K(t) \cos(3, K)(t). \quad (3)$$

But the macroscopic second order polarizations for a system of rod-like molecules are given by

$$P_x^{(2)}(t) = N \int p_3^{(2)}(t) \sin(3, x)(t) f(\Omega) d\Omega, \quad (4.1)$$

$$P_y^{(2)}(t) = N \int p_3^{(2)}(t) \sin(3, y)(t) f(\Omega) d\Omega, \quad (4.2)$$

$$P_z^{(2)}(t) = N \int p_3^{(2)}(t) \cos(3, z)(t) f(\Omega) d\Omega, \quad (4.3)$$

where N is the reorientable molecules density, $f(\Omega)$ is an angular density distribution function of the molecules and $d\Omega$ is a solid-angle element.

Thus, the non-zero second order susceptibilities terms are $\chi_{xzx}^{(2)}(t) = \chi_{xzy}^{(2)}(t) = \chi_{xxz}^{(2)}(t) = \chi_{xyz}^{(2)}(t)$, $\chi_{zyx}^{(2)}(t) = \chi_{yxz}^{(2)}(t) = \chi_{zyx}^{(2)}(t) = \chi_{yxz}^{(2)}(t)$, $\chi_{zxx}^{(2)}(t) = \chi_{zxy}^{(2)}(t) = \chi_{zxy}^{(2)}(t) = \chi_{zxy}^{(2)}(t)$, $\chi_{zzz}^{(2)}(t)$, which are explicitly expressed in the next equations:

$$\begin{aligned} \chi_{xzx}^{(2)}(t) &= N \int \beta_{333} \cos^2(3, x)(t) \cos(3, z)(t) f(\Omega) d\Omega \\ &= \frac{N}{2} \int \beta_{333} \sin^2(3, z)(t) \cos(3, z)(t) f(\Omega) d\Omega, \end{aligned} \quad (5.1)$$

$$\begin{aligned} \chi_{zyx}^{(2)}(t) &= N \int \beta_{333} \cos^2(3, y)(t) \cos(3, z)(t) f(\Omega) d\Omega \\ &= \frac{N}{2} \int \beta_{333} \sin^2(3, z)(t) \cos(3, z)(t) f(\Omega) d\Omega, \end{aligned} \quad (5.2)$$

$$\begin{aligned} \chi_{zxx}^{(2)}(t) &= N \int \beta_{333} \cos(3, z)(t) \cos^2(3, x)(t) f(\Omega) d\Omega \\ &= \frac{N}{2} \int \beta_{333} \cos(3, z)(t) \sin^2(3, z)(t) f(\Omega) d\Omega, \end{aligned} \quad (5.3)$$

$$\chi_{zzz}^{(2)}(t) = N \int \beta_{333} \cos^3(3, z)(t) f(\Omega) d\Omega, \quad (5.4)$$

or equivalently in the next equations:

$$\begin{aligned} \chi_{xzx}^{(2)}(t) &= \chi_{xzy}^{(2)}(t) = \chi_{xxz}^{(2)}(t) = \chi_{xyz}^{(2)}(t) \\ &= \frac{N\beta_{333}}{2} (\langle \cos \theta(t) \rangle - \langle \cos^3 \theta(t) \rangle), \end{aligned} \quad (6.1)$$

$$\begin{aligned} \chi_{zyx}^{(2)}(t) &= \chi_{yxz}^{(2)}(t) = \chi_{zyx}^{(2)}(t) = \chi_{yxz}^{(2)}(t) \\ &= \frac{N\beta_{333}}{2} (\langle \cos \theta(t) \rangle - \langle \cos^3 \theta(t) \rangle), \end{aligned} \quad (6.2)$$

$$\begin{aligned} \chi_{zxx}^{(2)}(t) &= \chi_{zxy}^{(2)}(t) = \chi_{zxy}^{(2)}(t) = \chi_{zxy}^{(2)}(t) \\ &= \frac{N\beta_{333}}{2} (\langle \cos \theta(t) \rangle - \langle \cos^3 \theta(t) \rangle), \end{aligned} \quad (6.3)$$

$$\chi_{zzz}^{(2)}(t) = N\beta_{333} \langle \cos^3 \theta(t) \rangle, \quad (6.4)$$

where ‘ $\langle \rangle$ ’ brackets mean an angular average, at each time, with its corresponding poling time-dependent density distribution function.

2.2. SHG intensity

The SHG intensity of a material depends on its ability for generating a beam of light with the double of the fundamental beam of light frequency, i.e. of the magnitude of its second order susceptibility, and also depends on the fundamental beam of light intensity. The intensity of the fundamental beam of light (I^ω) can be expressed in terms of its electric field vectors as

$$I^\omega = \frac{c\epsilon_0\pi}{2} \langle |E_x^\omega(t)|^2 + |E_y^\omega(t)|^2 + |E_z^\omega(t)|^2 \rangle, \quad (7)$$

where ‘ $\langle \rangle$ ’ brackets mean temporal average over one cycle of the electromagnetic wave, c is the speed of light in vac-

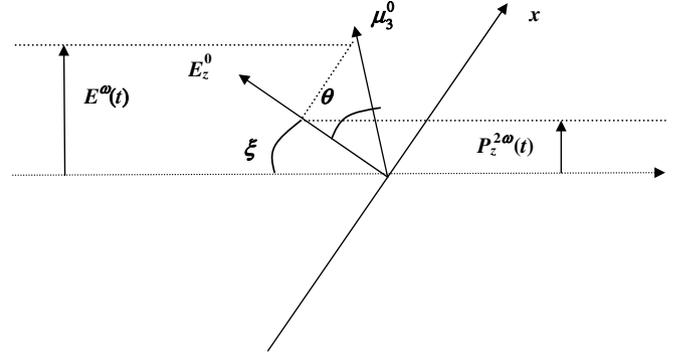


Fig. 2. Relation between macroscopic and microscopic coordinates, including a fundamental and a second harmonic generated beam of light.

uum, ϵ_0 is the vacuum permittivity and ω is the temporal frequency of the fundamental beam of light. If the fundamental beam of light was non-polarized then each one of its electric field vectors would take the next values

$$|E_x^\omega| = \sqrt{\frac{I^\omega}{c\epsilon_0\pi}} \cos \xi, \quad (8.1)$$

$$|E_y^\omega| = \sqrt{\frac{I^\omega}{c\epsilon_0\pi}}, \quad (8.2)$$

$$|E_z^\omega| = \sqrt{\frac{I^\omega}{c\epsilon_0\pi}} \sin \xi, \quad (8.3)$$

with ξ as the incidence angle of the fundamental beam of light on a material surface. Fig. 2 depicts the geometrical relation existing between a chromophore and the electric field vectors of light and the geometrical relation existing between a chromophore and a strong d.c. orienting field applied along the z axis (Corona field). θ is the angle between a chromophore permanent dipolar moment (μ_3^0 along the 3 axis) and a Corona field (E_z^0 along the z axis). ξ is the angle between the z axis and the beam of light propagation direction. x axis is on the plane of incidence of light and y axis is out of the plane of incidence. E^ω represents any fundamental light electric field vector. $P_z^{2\omega}$ represents the polarization field vector of the second harmonic light, polarization induced on a chromophore along the z axis.

In the same way the intensity of the second harmonic generated beam of light has to be directly proportional to the temporal average of the square of a material optically induced macroscopic second order polarization

$$I^{2\omega}(t) \propto \frac{c\epsilon_0\pi}{2} \langle |P_x^{(2)}(t)|^2 + |P_y^{(2)}(t)|^2 + |P_z^{(2)}(t)|^2 \rangle. \quad (9)$$

If the material is a system of reorientable rod-like molecules, just like some organically doped polymer-like materials, then the components of the optically induced macroscopic second order polarization take the values indicated by Eqs. (10), which come from the combination of Eqs. (4), (6) and (8).

$$P_x^{(2)}(t) = \frac{N\beta_{333}I^\omega}{c\epsilon_0\pi} (\langle \cos \theta(t) \rangle - \langle \cos^3 \theta(t) \rangle) (\cos \xi + 1) \sin \xi \cos \xi, \quad (10.1)$$

$$P_y^{(2)}(t) = \frac{N\beta_{333}I^\omega}{c\epsilon_0\pi} (\langle \cos \theta(t) \rangle - \langle \cos^3 \theta(t) \rangle) (\cos \xi + 1) \sin \xi, \quad (10.2)$$

$$P_z^{(2)}(t) = \frac{N\beta_{333}I^\omega}{c\epsilon_0\pi} \left[\langle \cos^3 \theta(t) \rangle \sin^2 \xi + (\langle \cos \theta(t) \rangle - \langle \cos^3 \theta(t) \rangle) \left(\frac{1}{2} + \cos \xi + \frac{\cos^2 \xi}{2} \right) \right] \sin \xi. \quad (10.3)$$

Now, with Eqs. (9) and (10) it is possible to obtain an expression for the SHG intensity in this kind of materials, as a function of the poling time

$$I^{2\omega}(t) \propto \left((\langle \cos \theta(t) \rangle - \langle \cos^3 \theta(t) \rangle)^2 (\cos \xi + 1)^2 (\cos^2 \xi + 1) + \left(\langle \cos^3 \theta(t) \rangle \sin^2 \xi + (\langle \cos \theta(t) \rangle - \langle \cos^3 \theta(t) \rangle) \left(\frac{1}{2} + \cos \xi + \frac{\cos^2 \xi}{2} \right) \right)^2 \right) (N\beta_{333}I^\omega)^2 \sin^2 \xi. \quad (11)$$

Poling time belongs to a larger interval than that of one electromagnetic wave cycle, in consequence poling time is not considered in the temporal average.

From Eq. (11) it is clear that SHG intensity is strongly dependent of the geometrical arrangement of the material rod-like molecules and it is clear that SHG intensity also depends on the incident angle of the fundamental beam of light.

2.3. Molecular angular average calculation

Eq. (11) poling time dependence comes exclusively from the polar angle poling time dependence for each reorientable molecule in the system. In Eq. (11) the polar angle only appears in the molecular angular averages. The molecular angular averages calculation is not a direct task due to their implicit poling time dependence. To calculate them in this work, we take into account the approach of our previous work [13], i.e. we assume that dynamics of each chromophore of the system under the influence of a strong d.c. electrostatic field (E_z^0) follows an over-damped harmonic oscillator behavior even for large initial angles. The next equation describes such kind of oscillator

$$\ddot{\theta} + 2\gamma\dot{\theta} + \omega_0^2\theta = \frac{I_{33}E_d}{\mu_3^0}, \quad (12)$$

where dots are derivatives with respect to the poling time, θ is the polar angle, γ is a damping constant related to the matrix–chromophore interactions, ω_0 is the natural oscillation frequency of the chromophore, and E_d is an extra field (non-related to the strong d.c. field), for example it can represent an effective field for the dipole–dipole electrostatic interactions among chromophores. ω_0 is related to the dipolar moment (μ_3^0) of the chromophore, to the main inertia moment (I_{33}) of the chromophore (in rod-like molecules their main inertia moment is almost aligned to its

dipolar moment), and to the effective local electric field (E) inside the material due to the strong d.c. field, as is shown in the next equation

$$\omega = \sqrt{\frac{\mu_3^0 E}{I_{33}}}. \quad (13)$$

The solution to Eq. (12) is the polar angle of one chromophore as function of the poling-time, but this solution depends on the chromophore initial conditions of angular position and angular speed.

The angular average of an arbitrary function which depends on the polar angle ($g(\theta(t))$) requires the knowledge of the chromophores distribution function time dependence. At the beginning we assume an angular uniform

distribution function, but for poling times different to zero we consider the next angular distribution function:

$$f(\Omega) = e^{\frac{\mu_3^0 E}{k_B T} \cos(\theta + \bar{\theta}(t))}, \quad (14)$$

with $k_B T$ the thermal energy of the system and $\bar{\theta}(t)$ as an angle at which the distribution function is centered for each poling time. $\bar{\theta}(t)$ is obtained as the inverse function of the function average over initial angular conditions with a uniform angular distribution, i.e. $\bar{\theta}(t) = g^{-1}(\langle g(\theta(t)) \rangle)$. Thus the required averages in Eq. (11) are numerically calculated for each poling time with Eqs. (15.2) and (15.3)

$$\langle g(\theta(t)) \rangle = \frac{\int_{\theta=0}^{\theta=\pi} 2\pi \sin \theta e^{\frac{\mu_3^0 E}{k_B T} \cos(\theta + \bar{\theta}(t))} g(\theta) d\theta}{\int_{\theta=0}^{\theta=\pi} 2\pi \sin \theta e^{\frac{\mu_3^0 E}{k_B T} \cos(\theta + \bar{\theta}(t))} d\theta}, \quad (15.1)$$

$$\langle \cos \theta(t) \rangle = \frac{\int_{\theta=0}^{\theta=\pi} 2\pi \sin \theta e^{\frac{\mu_3^0 E}{k_B T} \cos(\theta + \bar{\theta}(t))} \cos \theta d\theta}{\int_{\theta=0}^{\theta=\pi} 2\pi \sin \theta e^{\frac{\mu_3^0 E}{k_B T} \cos(\theta + \bar{\theta}(t))} d\theta}, \quad (15.2)$$

$$\langle \cos^3 \theta(t) \rangle = \frac{\int_{\theta=0}^{\theta=\pi} 2\pi \sin \theta e^{\frac{\mu_3^0 E}{k_B T} \cos(\theta + \bar{\theta}(t))} \cos^3 \theta d\theta}{\int_{\theta=0}^{\theta=\pi} 2\pi \sin \theta e^{\frac{\mu_3^0 E}{k_B T} \cos(\theta + \bar{\theta}(t))} d\theta}. \quad (15.3)$$

3. Experimental

The sample preparation for the materials used in this work to test this model has been previously reported [14]. The reorientable chromophore was Disperse Red 1 (DR1) functionalized in high concentration relative to the also functionalized molecule of Carbazole (SiK) with four different molar ratios: 1DR1:20Carbazole:1TEOS, 1DR1:10Carbazole:1TEOS, 1DR1:5Carbazole:1TEOS, 1DR1:3Carbazole:1TEOS. These spin-coated samples had a thickness around of 1.3 μm .

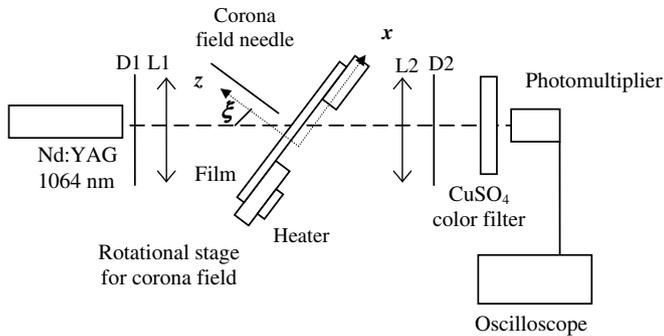


Fig. 3. Experimental setup scheme.

Disperse Red 1 (DR1) is a rod-like shape nanomolecule with 1.5 nm length, with a large permanent dipole moment along its main axis ($\mu_3^0 = 8.7 \text{ D}$ [1]), with a large second order hyperpolarizability constant along its main axis ($\beta_{333} = 125 \times 10^{-30} \text{ esu}$ [1]) and with its main inertia moment along the same axis ($I_{33} = 7.0 \times 10^{-43} \text{ kg m}^2$ [7]).

The second harmonic generation was measured employing the experimental setup depicted in Fig. 3. A Nanolase NP-10620-100 Nd:YAG non-polarized laser in the 1064 nm wavelength was employed as the source of the fundamental beam of light. Sol-gel film is heated and poled on a rotational stage. The orientation of the DR1 molecules in the sol-gel films was induced by a strong d.c. electrostatic field by means of the single-point Corona poling technique (needle-surface distance = 12 mm, voltage = +6 kV d.c., poling temperature = 120 °C). Corona field is applied perpendicular to the film main surface, the corona field determines the z axis of the macroscopic

$$I^{2\omega}(t) = C \left(\left(\langle \cos \theta(t) \rangle - \langle \cos^3 \theta(t) \rangle \right)^2 (\cos \xi + 1)^2 (\cos^2 \xi + 1) + \left(\langle \cos^3 \theta(t) \rangle \sin^2 \xi + \left(\langle \cos \theta(t) \rangle - \langle \cos^3 \theta(t) \rangle \right) \left(\frac{1}{2} + \cos \xi + \frac{\cos^2 \xi}{2} \right) \right)^2 \sin^2 \xi, \quad (16)$$

laboratory coordinate system. x axis of the macroscopic laboratory coordinate system is on the plane of incidence of light, and y axis is out of the plane of incidence. ξ is the angle between the propagation direction of the fundamental beam of light and the z axis. The emergent light is filtered with a CuSO_4 (copper sulfate) solution, so only the harmonic beam of light reaches the photomultiplier. The intensity of the harmonic beam of light is detected by a Hamamatsu H5784 photomultiplier and registered by a Tektronix TDS 3052B oscilloscope. D1 and D2 are diaphragms. L1 and L2 are lenses. It is necessary to mention here that the fundamental beam of light was non-polarized, and the detection of the second harmonic intensity was carried out with the non-polarized beam of light.

The measurements for the four different chromophore concentrations were carried out with a constant ξ angle (80°), and were plotted in a SHG intensity vs. poling time

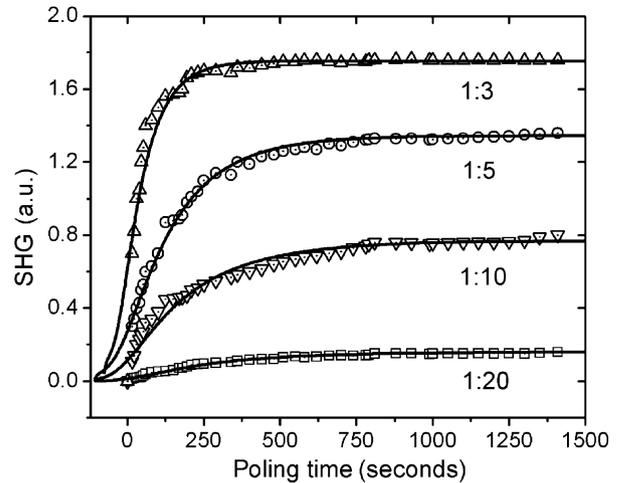


Fig. 4. Fittings to experimental data.

graph. Fig. 4 shows the obtained experimental data. The graph shows, in arbitrary units, the second harmonic generation intensity in the vertical axis and shows the poling time, in seconds, in the horizontal axis for films with four different DR1:Carbazole molar ratios. Symbols represents the experimental data and the solids lines are the fitting curves.

4. Results and discussion

The solid lines in Fig. 4 were plotted employing the next equation

which is the same one that Eq. (11) but with an explicit proportionality factor C . The angular averages in Eq. (16) were carried out with Eqs. (15). The fitting parameters employed for each curve are listed in Table 1. In all cases, a typical effective local electric field (E) with a value equal to $6.01 \times 10^8 \text{ V/m}$ was considered for this kind of sol-gel films.

Table 1
Parameters used for the best fitting to the experimental results obtained for each studied film

DR1:Carbazole:TEOS molar ratios	γ (s^{-1})	C	Time shift (s)	E_d (V/m)
1:20:1	0.45×10^{25}	0.38	110	0
1:10:1	0.34×10^{25}	1.84	110	0
1:5:1	0.22×10^{25}	6.08	110	3.999×10^8
1:3:1	0.11×10^{25}	17.06	80	6.056×10^8

The fittings require two fitting parameters: the over-damping constant (γ) and the proportionality factor C (for the samples with the two lowest chromophores concentration) or the over-damping constant (γ) and the effective field of chromophore–chromophore interactions (E_d) (for the samples with the two largest chromophores concentration). Additionally, Table 1 includes a time shift value. This time shift represents a displacement in time for the fittings, i.e. fittings obtained from Eq. (16) have their temporal origin shifted with respect to the experimental data, due to the assumption of the harmonic movement for any chromophores initial angular position (the chromophores with the largest initial polar angles have a different angular speed than the other chromophores). It is remarkable that three of the four samples have the same time shift value.

In all cases the asymptotic behavior of the samples is well described by the model. The SHG intensity asymptotic value reached by the samples depends on two parameters of Table 1: the chromophore concentration and the effective field of chromophore–chromophore electrostatic interactions (E_d). According with Eq. (11), the intensity ratio between two samples must be proportional to the square of their concentration ratio. For the sample with the lowest DR1 concentration, the chromophore–chromophore interactions were not considered. For the sample with the 1:10:1 molar ratio, the concentration ratio with former was 2, and then it was expected a SHG intensity ratio equal to 4, that agrees with the C ratio from Table 1: $1.84/0.38 = 4.8$. In consequence $E_d = 0$ for this second concentration, too. For the sample with the 1:5:1 molar ratio, the concentration ratio with the lowest one was 4, and then it was expected a SHG intensity ratio equal to 16, but the SHG intensity ratio was $1.36/0.16 = 8.5$, too low, in consequence there are chromophore–chromophore interactions and E_d becomes into the fitting parameter. Similarly, for the sample with the 1:3:1 molar ratio concentration the chromophore–chromophore interactions result quite high (E_d has a similar magnitude than the typical effective local electric field (E) inside the film).

With respect to the over-damping constant (γ), which represents the matrix–chromophore interactions, it is interesting to note that its values become lower as the DR1 concentration increases. This situation can be clearly observed in Fig. 4, because as the samples increases their DR1 concentration they reach faster their maximum asymptotic SHG intensity value.

5. Conclusions

Second harmonic generation experimental results were fitted as a function of the poling time, in SiO₂:DR1 side-chain doped sol–gel films for several DR1 chromophore concentrations and a non-polarized fundamental beam of light. This macroscopic response depends on the orientation and interaction of these nanoscopic molecules. An increment in the SHG intensity beam of light was observed as the chromophore concentration increases, however, such increment is not linear due to an electrostatic interaction among the chromophore permanent dipoles, and in consequence their optimal orientation inside the film is not achieved. The model fittings make a good description of the experimental results.

Acknowledgments

This work has been supported by the following grants: ECOS M02-P02, PUNTA-UNAM, CONACyT 43226, DGAPA-UNAM IN111902, UCMEXUS and CONACyT-NSF.

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