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N. Pereda

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# Second harmonic generation in planar samples of ferroelectric liquid crystals

N. PEREDA, C. L. FOLCIA, J. ETXEBARRIA*
Departamento de Física de la Materia Condensada, Facultad de Ciencias,
Universidad del País Vasco, 48080 Bilbao, Spain

and J. ORTEGA
Departamento de Física Aplicada II, Facultad de Ciencias,
Universidad del País Vasco, 48080 Bilbao, Spain

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Non-linear optical properties of ferroelectric liquid crystals are often studied through second harmonic generation measurements on homeotropic cells. However, it is not always possible to prepare well aligned unwound samples in the homeotropic geometry, especially when dealing with photopolymers or tightly twisted SmC* materials. Here we analyse the problem of extracting the second order susceptibility coefficients $d_{ij}$ from second harmonic generation measurements performed on planar samples. Several experimental configurations of polarizer, sample and analyser dispositions are proposed to deduce the complete $d_{ij}$ tensor together with the dispersion of the refractive indices and the birefringence both at the fundamental and second harmonic frequencies. Measurements carried out on planar and homeotropic cells for a classical non-linear optical material (W314) are presented and the resulting $d_{ij}$ coefficients are compared.



## 1. Introduction

In the last few years the design of ferroelectric liquid crystals (FLCs) with large molecular second order hyperpolarizabilities for non-linear optics has attracted much attention due to their applicability in the fabrication of ultra-fast electro-optic modulators [1–6]. More recently, the interest has been focused on the synthesis of photo-polymers derived from FLC monomers [7, 8]. These high molecular mass compounds have a large polar order and at the same time are mechanically more stable than their low molecular mass counterparts.

Non-linear optical (NLO) properties of low and high molecular mass FLCs are often studied through second harmonic generation (SHG) measurements. In this process the NLO material converts light at frequency $\omega$ into light at $2\omega$. The efficiency is basically determined by the second order susceptibility tensor ($d_{ij}$). In the case of the ferroelectric SmC* phase, this tensor has four independent coefficients, whose determination fully characterizes the NLO properties of the material.

The homeotropic geometry is the most frequently used in SHG studies. In this configuration, as is well known, the phase matching (PM) condition is usually achieved at an angle of incidence easily accessible experimentally (0°–30°) for the ee-o conversion [9, 10] (here e and o stand for extraordinary and ordinary polarizations, respectively). From measurements around the PM peak, three non-linear coefficients ($d_{21}$, $d_{23}$, $d_{25}$) can be deduced, while the fourth coefficient ($d_{22}$) (usually optimized in the synthesis process) can be obtained from measurements for the oo-o conversion [11].

However, although homeotropic samples seem to be the most appropriate for SHG investigations, they are not always practicable. In fact, we have observed that electrohydrodynamic instabilities appear in several homeotropically aligned materials when applying electric fields in order to unwind the helix characteristic of the SmC* phase. Such a phenomenon occurs especially when dealing with photo-polymers or tightly twisted SmC* materials and is presumably due to the existence of large electric field gradients at the borders of the metallic spacers (which also operate as electrodes) in those homeotropic samples. Therefore, we consider it of interest to investigate other geometries, in particular the planar one. In fact, several studies on SHG on planar samples have already been published [8, 12–15]. Nevertheless, most of these reports are incomplete (an exception is ref. [8]) in the sense that the authors have limited themselves simply to obtaining an insight of the properties of the NLO materials or extracting partial

*Author for correspondence.

information. In this work we have combined several configurations and developed a method which permits us to obtain the complete ($d_{ij}$) tensor. Our method has been applied to study a thin planar sample of a calamitic compound (W314) with a relatively high NLO behaviour [16]. The results will be compared with those obtained using the usual homeotropic configuration.

## 2. SHG in FLC cells

In this section we will investigate the second order non-linear optical behaviour of FLCs when they are illuminated with light at frequency $\omega$. As a result of the macroscopic symmetry characteristic of the SmC* phase (a twofold axis parallel to the spontaneous polarization), the second order susceptibility tensor is given by:

$$(d_{ij}) = \begin{bmatrix} 0 & 0 & 0 & d_{25} & 0 & d_{21} \\ d_{21} & d_{22} & d_{23} & 0 & d_{25} & 0 \\ 0 & 0 & 0 & d_{23} & 0 & d_{25} \end{bmatrix} \quad (1)$$

where Kleinman conditions are assumed to be valid. The tensor is referred to a coordinate system $(x, y, z)$ whose $y$-axis is along the polarization $\mathbf{P}_s$ and the $z$-axis is parallel to the optic axis (the direction of the molecular director under the uniaxial approximation) (see figure 1).

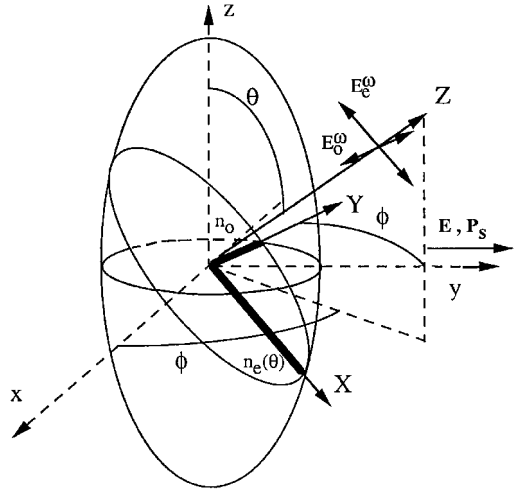


Figure 1. Laboratory ($X, Y, Z$) and sample ($x, y, z$) frames used to study the SHG process in FLCs. The sample is assumed to behave optically as a positive uniaxial medium, whose index ellipsoid is schematized in the figure. The molecular director is along the optic axis, which is parallel to $z$. The spontaneous polarization $\mathbf{P}_s$ has the direction of the $y$-axis. The light is incident along the negative direction of the $Z$-axis with polar and azimuthal angles $\theta$ and $\phi$ with respect to the sample frame. The (linear) polarization eigenmodes of the incident light are taken along the X (extraordinary) and Y (ordinary)-axes.

The amplitude of the SH field generated by a NLO material can be expressed as [17]:

$$E_I^{2\omega}(L) = \frac{-i\omega}{cn_I^{2\omega}} \sum_{J,K} d_{IJK} E_J^\omega E_K^\omega \frac{\exp(i\Delta k_{IJK} L) - 1}{i\Delta k_{IJK}}$$

$$\Big/ I, J, K = X, Y \quad (2)$$

for incoming linearly polarized light of frequency $\omega$ propagating along $Z$. $X, Y$ are the directions of the eigenmodes of polarization in the crystal (see figure 1), $L$ is the interaction length and $d_{IJK}$ are the coefficients of the second order susceptibility tensor referred to this frame, each of them being a linear combination of the different $d_{ij}$ parameters of equation (1). $\Delta k_{IJK} = k_I^{2\omega} - (k_J^\omega + k_K^\omega)$ is the phase mismatch, where $k_J^\omega = \omega n_J^\omega / c$, $k_I^{2\omega} = 2\omega n_I^{2\omega}/c$ are the wave vectors of the eigenmodes with electric field amplitudes $E_J^\omega$ and $E_I^{2\omega}$ respectively.

We will assume that materials behave as positive uniaxial media and show normal dispersion. In this case the ordinary and extraordinary rays are along $Y$ and $X$ directions, respectively. They propagate with refractive indices $n_o$ and $n_e(\theta)$, where $n_e(\theta)$ is given by:

$$\frac{1}{[n_e(\theta)]^2} = \frac{\cos^2\theta}{n_o^2} + \frac{\sin^2\theta}{n_e^2} \quad (3)$$

and $n_o$ and $n_e$ are the ordinary and extraordinary indices of the material.

In general, three different terms contribute to equation (2) (as $d_{IJK} = d_{IKJ}$ and $\Delta k_{IJK} = \Delta k_{IKJ}$). Since the SHG signal is obtained by squaring the field amplitude, it will be described by a very complex expression. All the three $d_{IJK}$ parameters, together with the $n_o$ and $n_e$ indices, both at the fundamental and SH frequency, appear mixed. Some terms in equation (2) may fulfil the PM condition both for type I [10] (ee-o) or type II [15] (eo-o) conversions, for certain configurations of $\mathbf{E}^\omega$ and $\mathbf{E}^{2\omega}$. In such situations, it can be considered that the effect of these terms is predominant with respect to the others, and equation (2) is greatly simplified. On the other hand, in a general case it is difficult to extract the non-linear $d_{ij}$ coefficients and therefore it is important to investigate special configurations where the SH signal expression is simplified.

The most suitable dispositions are those in which one single term appears in equation (2). This can be achieved when the director is maintained parallel or perpendicular to the plane of incidence during the whole experiment. Typically this is attained by rotating the liquid crystal cell around a vertical axis parallel or perpendicular to the molecular director while keeping fixed the incident laser beam. The p and s polarizations are then obtained

with horizontal and vertical polarizers, respectively. These polarizations correspond to the o and e eigenmode directions if the director is vertical and to e and o when it is horizontal. Under such conditions the SH power $P^{2\omega}$ can be expressed by the standard formula:

$$P_{\mathrm{I}}^{2\omega} = \frac{8\omega^2 d_{\mathrm{IJJ}}^2 (t_{\mathrm{J}}^{\omega})^4 (t_{\mathrm{I}}^{2\omega})^2}{\varepsilon_0 c^3 (n_{\mathrm{J}}^{\omega})^2 n_{\mathrm{I}}^{2\omega} A} L^2 \times \mathrm{sinc}^2\left(\frac{\Delta k_{\mathrm{IJJ}} L}{2}\right)(P_{\mathrm{J}}^{\omega})^2 \qquad \mathrm{I, J = p, s} \tag{4}$$

where $d_{\mathrm{IJJ}}$ is the so-called $d_{\mathrm{eff}}$ coefficient for each particular configuration, $A$ is the beam area, $P^{\omega}$ is the power of the incident light and $t_{\mathrm{I}}^{\omega}$, $t_{\mathrm{I}}^{2\omega}$ are the standard Fresnel transmission factors for the I-polarized light with frequencies $\omega$ and $2\omega$ [18].

In the usual geometry with homeotropic cells, the director is set horizontal and the SH light is s-polarized (ordinary) (see figure 2). For the oo-o configuration, equation (4) gives rise to Maker fringe profiles, while at the ee-o configuration the PM condition is achieved when $n_{\mathrm{e}}^{\omega}(\theta) = n_{\mathrm{o}}^{2\omega}$, which occurs for a polar angle such that:

$$\sin^2\theta \approx \frac{\Delta n_{\mathrm{d}}}{\Delta n^{\omega}} \tag{5}$$

where $\Delta n^{\omega} = n_{\mathrm{e}}^{\omega} - n_{\mathrm{o}}^{\omega}$ is the birefringence at the fundamental frequency and $\Delta n_{\mathrm{d}} = n_{\mathrm{o}}^{2\omega} - n_{\mathrm{o}}^{\omega}$ accounts for the material dispersion. The corresponding $d_{\mathrm{eff}}$ coefficients involved in each case are:

$$d_{\mathrm{eff}}(\mathrm{ee\text{-}o}) = d_{21}\cos^2\theta + d_{23}\sin^2\theta + d_{25}\sin 2\theta \tag{6}$$

$$d_{\mathrm{eff}}(\mathrm{oo\text{-}o}) = d_{22}. \tag{7}$$

As can be seen, the four coefficients $d_{ij}$ of the second order susceptibility tensor are present, making their determination relatively simple and accurate if we follow the procedure described in previous studies [19, 20].

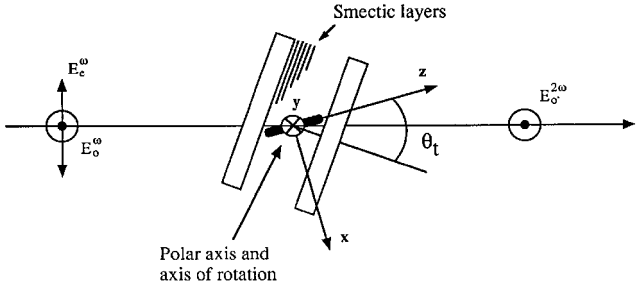


Figure 2. Typical experimental geometry used to study the SHG process in homeotropic samples. The cell is rotated about the polar axis and the SH signal is measured as a function of the angle of incidence of the incoming light. For ee-o conversion type I PM can be achieved.

We will now investigate several planar dispositions which are suitable for SHG studies. In particular, we propose five different configurations in order to obtain the complete $d_{ij}$ tensor, as well as the dispersion of the refractive indices and the birefringence, both at the fundamental and SH frequencies.

In the two first measurements, M1 and M2 (see figure 3), the cell, arranged in such a way that the director is vertical (M1) or horizontal (M2), is rotated around a vertical axis. In both cases, the polarizer and analyser are set horizontally. One single term contributes then to the SH signal and therefore $P^{2\omega}$ is described by equation (4).

In particular, the SH power for each case can be expressed as a function of the angle of refraction $\theta_{\mathrm{r}}$ as:

$$P^{2\omega}(\mathrm{M1}) \propto (3d_{21}\cos^2\theta_{\mathrm{r}}\sin\theta_{\mathrm{r}} + d_{22}\sin^3\theta_{\mathrm{r}})^2 \times \frac{\sin^2\left(\frac{2\pi}{\lambda}\Delta n_{\mathrm{d}} L\right)}{\left(\frac{2\pi}{\lambda}\Delta n_{\mathrm{d}}\right)^2} \tag{8}$$

$$P^{2\omega}(\mathrm{M2}) \propto (3d_{23}\cos^2\theta_{\mathrm{r}}\sin\theta_{\mathrm{r}} + d_{22}\sin^3\theta_{\mathrm{r}})^2 \times \frac{\sin^2\left(\frac{2\pi}{\lambda}\Delta n_{\mathrm{d}} L\right)}{\left(\frac{2\pi}{\lambda}\Delta n_{\mathrm{d}}\right)^2} \tag{9}$$

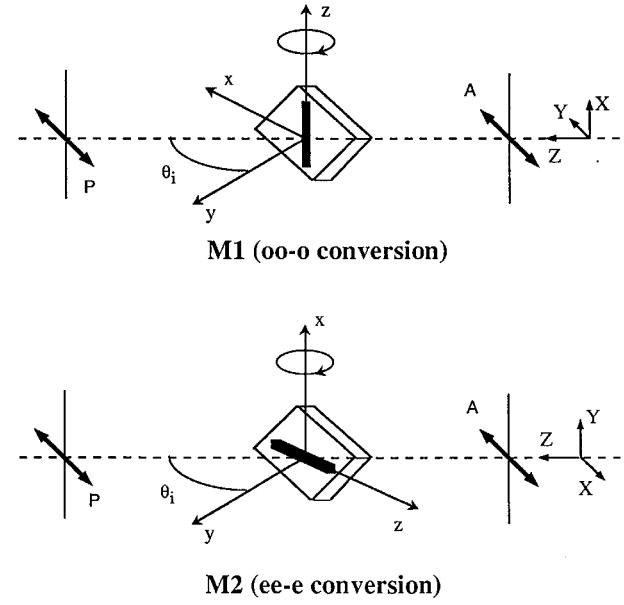


Figure 3. M1 and M2 dispositions used to study the SHG in planar samples. In both cases $P^{2\omega}$ is measured as a function of the angle of incidence. The polarizer (P) and the analyser (A) are set horizontally (pp-p configuration). M1 corresponds to an oo-o conversion and in M2 the process is described as ee-e.

where $\lambda$ is the fundamental beam wavelength and we have assumed that the birefringence both at the fundamental and SH frequency is the same.

On the other hand, for small interaction lengths ($L \leqslant 3-4\ \mu m$) and typical values for the dispersion $\Delta n_d$ in FLCs we have:

$$\sin^2\left(\frac{2\pi\Delta n_d L}{\lambda}\right) \approx \left(\frac{2\pi\Delta n_d L}{\lambda}\right)^2 \qquad (10)$$

and, thus, the signal will be almost independent of the refractive indices. Once the experimental points are corrected from the corresponding Fresnel transmission factors, see equation (4), we can perform a fit to equations (8) and (9) in order to extract $d_{21}$, $d_{23}$ and $d_{22}$. It is to be noted that another 6 similar combinations (vertical–horizontal) of polarizer–director–analyser dispositions (different from M1 and M2) also give rise to an expression for $P^{2\omega}$ of the type of equation (4). However, it can be shown that they are not relevant, because either they give redundant information or the corresponding SHG signal is very small or even null.

We have instead considered other configurations (M3–M5) (see figure 4) which are useful in refining the above $d_{ij}$ results, as well as for obtaining information about $d_{25}$, the dispersion and birefringence. In M3 and M4 the analyser is fixed horizontally and the cell is oriented in such a way that the director is vertically (M3) or horizontally (M4) arranged. In the last disposition (M5), the director is horizontal while the analyser is vertically set. In the three cases, the incidence is fixed at an angle different from zero and the SH signal is measured as a function of the polarizer angle. In all these cases the functional dependence of $P^{2\omega}$ is very complex because a lot of material parameters are involved. In principle, the equations can be greatly simplified when the cell thickness is very small ($L \leqslant 1\ \mu m$), since the phase factor $\exp(i\Delta kL)$ in equation (2) can be approximated to

$$\exp(i\Delta kL) \approx 1 + i\Delta kL \qquad (11)$$

leading to expressions for $P^{2\omega}$ with no dependence on the refractive indices. However, we have checked that for interaction lengths typical for thin cells ($L \geqslant 2\ \mu m$), approximation (11) is not justified and the general expression of equation (2) must be considered. A simple fit to extract the unknown coefficients is then not possible. The only feasible way to analyse these data is through simulations, using a trial and error method, until the resulting profiles are consistent with the experimental points, both in shape and in size. Consequently we have devised a program that, once the measurement conditions and the relevant coefficients have been inserted, performs a simulation of the SHG signal as a function of the polarization angle of the incoming light. In contrast to the fits in M1 and M2, we are forced in these other cases to insert the corresponding Fresnel factors into each field amplitude, since it is impossible to correct the experimental points because the different Fresnel factors appear admixed.

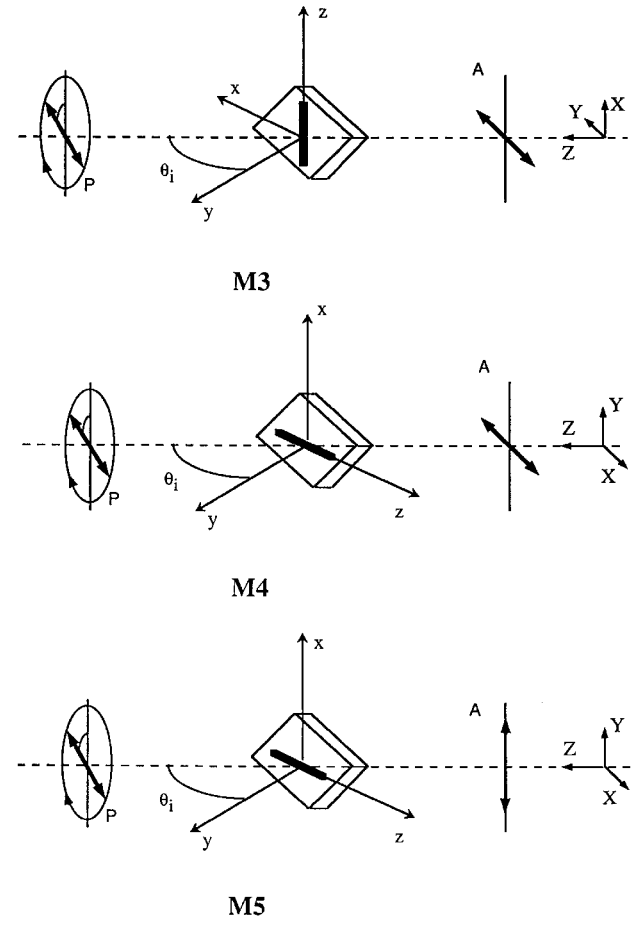


Figure 4. M3–M5 dispositions used to study the SHG process in planar samples. The angle of incidence is fixed ($\theta_i = 20°$ in our measurements) while the SH signal is measured as a function of the angle of the polarizer.

## 3. Experimental results

We have applied our method to a FLC compound (W314) especially designed by Walba *et al.* [1] for NLO applications. A commercial planar cell (EHC) was used. The thickness (3.0 μm) was determined by means of an interferometric technique. A good alignment of the smectic layers was obtained in the SmC* phase by slow cooling from the isotropic phase under a square-wave voltage of 20 V and 5 Hz. The cell was mounted on a temperature-controlled rotating stage with optical access. The temperature was maintained fixed at 75°C ($T - T_c \approx 15°C$) for all the measurements. A standard technique was used in our SHG studies [20]. The

fundamental light beam came from a Q-switched Nd:YAG laser (wavelength $\lambda = 1064$ nm, pulse width 6 ns and repetition rate 5 pulses $s^{-1}$), with a peak intensity at the sample position of about 10 MW $cm^{-2}$. The second harmonic intensity was detected by a photomultiplier after passing through an IR cut filter, a green glass filter and an interference filter. Another photomultiplier at a reference branch was used to detect simultaneously the SH light generated by a $LiNbO_3$ crystal. The signal from the sample was divided by the signal from the reference crystal in order to compensate for the laser pulse spreading and intensity fluctuations. A $y$-cut quartz sample ($d_{11} = 0.4$ pm $V^{-1}$) was used as a standard for calibration purposes.

We performed the measurements in the five configurations (M1–M5) described in the previous section. The results are shown in figures 5 and 6. The experimental points corresponding to M1 and M2 have been divided by the Fresnel transmission factors, while for M3–M5 these factors are included in the simulation profiles, as explained in the previous section.

The $d_{ij}$ coefficients were deduced by processing the data in two steps. First we used M1 and M2 to get approximate values for $d_{21}$, $d_{22}$ and $d_{23}$ by fitting the experimental points to equations (8) and (9), assuming a typical value for $\Delta n_d \approx 0.03$. The preliminary values obtained in this way were:

$$d_{21} = 0.06 \pm 0.01 \text{ pm V}^{-1}$$
$$d_{23} = 0.12 \pm 0.02 \text{ pm V}^{-1} \quad (12)$$
$$d_{22} = 0.3 \pm 0.1 \text{ pm V}^{-1}.$$

The error for each coefficient was estimated by taking into account the error from the fits, as well as the scattering of the $d_{ij}$ values when a reasonable range for $\Delta n_d$ ($|\Delta n_d| \leqslant 0.05$) was considered in equations (8) and (9).

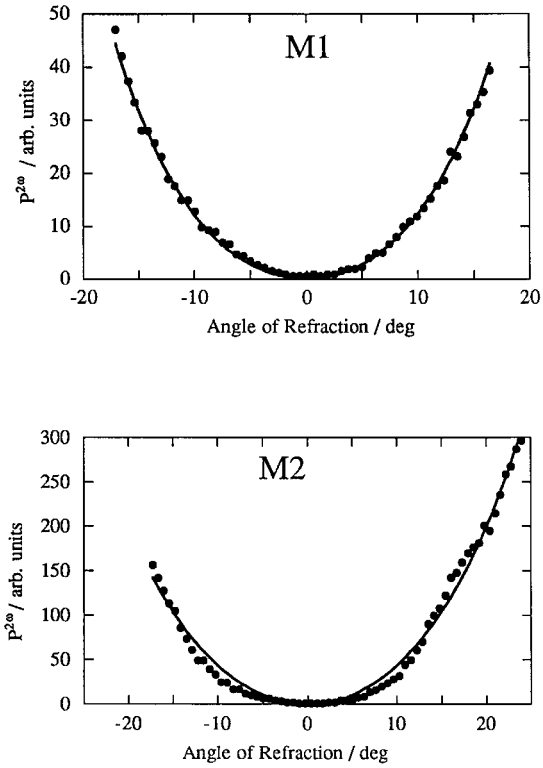


Figure 5. SH intensity as a function of the internal angle of refraction for the M1 and M2 geometries. The continuous lines correspond to the fits to equations (8) and (9).

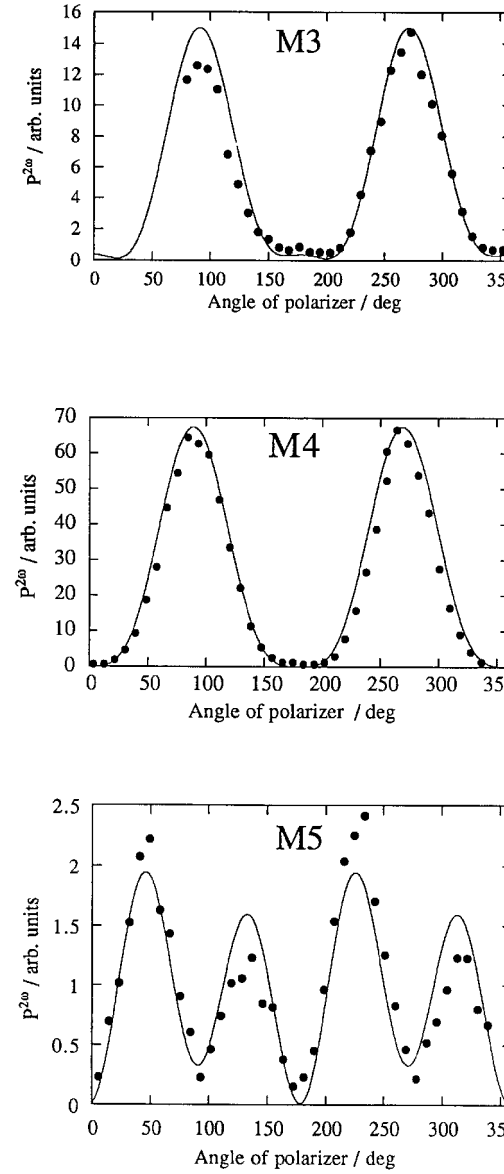


Figure 6. SH intensity as a function of the angle of the polarizer for the M3–M5 arrangements. The continuous lines correspond to the resulting theoretical curves with the parameters of equations (13).

For the second part of the data analysis, we turned to the simulation program in order to reproduce the experimental M3–M5 profiles. In the M3 configuration, all the $d_{ij}$ coefficients as well as the refractive indices are involved in the SHG signal, but it is $d_{21}$ which gives the main contribution. We could observe that the shape of the minimum was especially sensitive to small variations of $d_{25}$ and $\Delta n_d$. In M4 all the parameters again appear mixed, and in this case $d_{23}$ is the dominant coefficient. We checked that changes in $d_{22}$ and $\Delta n_d$ modify the curvature of the maxima and minima, while variations of the rest of the parameters do not substantially alter the profile. The last measurement (M5) permits us to distinguish between the birefringence at $\omega$ and $2\omega$. Moreover, in this case the only coefficients that participate in the SHG process are $d_{21}$ and $d_{25}$. From the results in this configuration we could deduce that $d_{25}$ is very small.

We worked simultaneously with the three configurations, changing slightly the original input for each simulation, in order to obtain a consistent relation between the experimental points and the simulation curves. After several iterations, a quite good agreement was reached, as is shown in figures 5 and 6. The values of the parameters which best reproduce the five experimental profiles are:

$$\begin{aligned} d_{21} &= 0.060 \pm 0.003 \text{ pm V}^{-1} & \Delta n^{\omega} &= 0.16 \pm 0.01 \\ d_{23} &= 0.120 \pm 0.005 \text{ pm V}^{-1} & \Delta n^{2\omega} &= 0.18 \pm 0.01 \\ d_{22} &= 0.3 \pm 0.1 \text{ pm V}^{-1} & \Delta n_d &= 0.02 \pm 0.01 \\ d_{25} &= -0.02 \pm 0.01 \text{ pm V}^{-1} \end{aligned} \tag{13}$$

The errors of the parameters are just estimates which were roughly calculated by examining globally the deviations of the experimental points from the theoretical curves in the five measurements M1–M5.

We now compare these results with those obtained from studies on homeotropic samples. Following the usual technique and using a 60 μm thick sample homeotropically aligned with CTAB and unwound by an in-plane electric field of 100 V mm$^{-1}$, we obtained $d_{\text{eff}} = 0.09$ pm V$^{-1}$ at the PM peak. On the other hand, from the data of equation (13) and taking a tilt angle $\theta_t = 30°$, a value of $d_{\text{eff}} = 0.08$ pm V$^{-1}$ is deduced from equation (6). As can be seen, both results are in good agreement. Unfortunately, the quality of the measurements on the homeotropic geometry did not allow us to get $d_{21}$, $d_{23}$ and $d_{25}$ separately, although we found that the data of equation (13) reproduced very well both the shape and size of the whole experimental curve. It is to be noted that the $d_{\text{eff}}$ value quoted in early studies (0.23 pm V$^{-1}$) [16] does not correspond with our results. This discrepancy could be attributed to the difference in temperatures at which the measurements were carried out (75 and 60°C respectively), although the disagreement is probably still large. One possible explanation of an overestimation of the $d_{ij}$ coefficients is neglect of the influence of Fresnel factors in the data process (especially if the reference sample is strongly reflecting, as is the case in the work of ref [16]), but with the available information it is not possible to establish the origin of the difference. Anyway the coincidence between our results on planar and homeotropic samples seems to give some confidence in the reliability of the present measurements.

In summary, we have studied the SHG process of a planarly aligned FLC sample. The relevant non-linear and linear optical parameters of the material have been deduced from measurements of $P^{2\omega}$ in different configurations. The results are consistent with those obtained using homeotropic cells. The present measuring technique can be useful for SHG studies on materials with high viscosity such as NLO FLC polymers.

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