Fine structure in spatial self-phase modulation patterns: at a glance determination of the sign of optical nonlinearity in highly nonlinear films

A fast and simple method to determine the sign of the nonlinear coefficient $n_2$ in films with high nonlinearity is presented. The method is similar to z-scan but does not require an aperture. It exploits the effect of spatial self-phase modulation and the role of wavefront curvature that leads to a specific far field pattern that is dependent on the sign of $n_2$ and on the sample position. The method can be considered a 'visualization' of the z-scan for highly nonlinear films. Application to nematic liquid crystals under different experimental conditions confirms the results predicted by theory.

3.1 Introduction

In this chapter we discuss a fast and easy method we devised to determine the sign of optical nonlinearity in thin nonlinear films \cite{1}. Liquid crystals were used as samples for measurement. In general, materials that simultaneously possess some properties typical of liquids and some others typical of solids are termed liquid crystals. These are materials that show mesomorphic (of intermediate form) states, which are aggregation states that lie in between the solid phase and the liquid phase \cite{2}. Most of the time liquid crystals show anisotropic physical properties. Liquid crystals are mainly divided into two categories, namely, thermotropic liquid crystals
and lyotropic liquid crystals, depending on the phase inducing factors. In thermotropic liquid crystals, changing the temperature of the system induces the phase transitions. Majority of the liquid crystalline compounds fall in this category. Lyotropic liquid crystalline phases are shown by amphiphilic molecules (molecules which have a hydrophilic and hydrophobic part) dissolved in a solvent. In this case changing the relative concentration of various components of the molecule brings about the phase transitions {3}.

In a mesomorphic compound, the chemical composition produces an orientational order of the molecules. The liquid crystalline phases are generally distinguished by the degree of molecular order they present. In the ‘nematic phase’ there exists an orientational order, but no positional order. On the average, molecules align their axes along a preferred direction and hence rotational symmetry around this direction can be found. This will in turn become a locally uniaxial phase. The ‘smectic phase’ is characterized by a layered structure. This is more ordered than the nematic phase. If a material posesses both nematic and smectic phases, the latter will appear at lower temperatures. The ‘cholesteric phase’ is generally considered a chiral nematic phase where the preferred molecular direction is subjected to helical distortion. This phase shows some similarities with the smectic phase as a layered structure sometimes can be identified {4}.

Figure 3.1: Nematic, smectic and cholesteric phases exhibited by liquid crystals.
In all liquid crystals, a preferred local molecular orientation can be found and its direction is usually indicated by a unit vector \( \mathbf{n} \) called “director”. This represents the average direction in a small volume, containing many molecules to make a meaningful averaging. The three commonly used director alignments determined by the boundary conditions of the liquid crystals are homeotropic (HOM), planar (PLAN) or homogenous, and hybrid alignments.

Figure 3.2: Homeotropic, Planar, and Hybrid alignments of liquid crystals.

In homeotropic alignment the director is normal to the surface. In the planar sample, the director alignment is tangential to the boundary. The planar and the homeotropic samples will have uniform alignment in the whole volume. On the contrary, the hybrid alignment is a distorted configuration created by homeotropic orientation at one boundary and planar orientation at the other boundary. Such a hybrid liquid crystal sample is referred to as HAN cell (Hybrid Aligned Nematic). Some other configurations used are the twisted and supertwisted ones, which are common in display applications \(^5\). These are often obtained by doping the nematic liquid crystal with a chiral compound (such as a cholesteric compound) and using planar alignments at both the boundaries. The experiments described in this chapter are done on nematic liquid crystal samples of homeotropic and planar alignment.

3.2 Spatial self-phase modulation in thin nonlinear films

When a Gaussian laser beam passes through a nonlinear medium, it may exhibit a characteristic diffraction ring pattern resulting from spatial self-phase modulation (SPM) \(^6\). In general in a liquid crystal medium, the laser beam can induce an unusually large refractive index modulation due to molecular
reorientation \( \{2\} \). The resulting spatial self-phase modulation on the laser beam usually yields a diffraction pattern in the form of many concentric rings.

![Figure 3.3: SPM rings observed in pentyl-cyanobiphenyl (5CB) nematic liquid crystal.](image)

Self-phase modulation is a consequence of the difference in nonlinear refractive index at different radial positions of a beam due to its non-uniform spatial intensity profile. The variation of the refractive index not only produces different light speeds leading to self-focusing or self-defocusing \( \{7\} \); but also induces changes in the optical path of each part of the beam. Since the manifestation of SPM is closely related to the temporal and spatial behavior of a light beam, the analysis of the patterns produced by a thick sample becomes very complicated. This is mainly due to the fact that SPM and self-focusing (or self-defocusing) effects can generally coexist in a thick sample and the theoretical treatment becomes rather complex. For the sake of simplicity, we consider only thin nonlinear samples for the analysis in this chapter.

In the case of a thin sample, where self-focusing and self-defocusing effects can be neglected, we can describe the SPM phenomenon caused by a Gaussian beam as diffraction of light through a circular aperture, which introduces a phase shift depending on the local light intensity. The circular aperture corresponds to the beam width on the sample, while the phase shift profile is due to the nonlinear
response of the medium. Following the classical theory of Fraunhofer diffraction \cite{8} we can write the light field in rectangular coordinates as,

\[ U(P) = C \int_A E_i e^{-ik(p\xi + q\eta)} d\xi d\eta \] - (3.1)

where \((p,q)\) is the coordinate of a point \(P\) in the diffraction pattern, \((\xi,\eta)\) is the coordinate of a typical point in the aperture and \(C\) is a constant. \(E_i\) is the incident electric field and \(k\) is the wave vector of the incident electromagnetic radiation.

For a circular aperture, it is more convenient to use polar coordinates to represent the light field. Let \((\rho,\theta)\) be the polar coordinates of a typical point in the aperture. We can write,

\[ \rho \cos \theta = \xi, \quad \rho \sin \theta = \eta \] - (3.2)

and let \((\omega,\psi)\) be the coordinates of a point \(P\) in the diffraction pattern referred to the geometrical image of the source. Defining

\[ \omega \cos \psi = p, \quad \omega \sin \psi = q \] - (3.3)

it follows that \(\omega = \sqrt{p^2 + q^2}\) is the sine of the angle which the direction \((p,q)\) makes with the central direction \(p=q=0\). If \(a\) is the radius of the circular aperture, the diffraction integral now can be written as,

\[ U(P) = C \int_0^{2\pi} \int_0^a E_i(\rho) e^{-ik\omega \cos(\theta - \psi)} \rho d\theta d\rho \] - (3.4)

For an incident Gaussian beam, the electric field can be written as \cite{8},

\[ E_i = E_0 e^{-\left(\frac{r^2}{w^2}\right)} e^{-\left(\frac{ikr^2}{2R}\right)} \] - (3.5)

where \(r\) is the radial distance of a point on the wave front from the center of the beam, \(w\) is the beam width, and \(R\) is the radius of curvature of the wave front. As we can see from the equation, for a Gaussian beam, the intensity is a function of \(r\). Hence the nonlinear phase shift introduced at the aperture should also be a
function of \( r \). If this nonlinear phase shift can be represented by \( \varphi_{\text{NL}}(r) \), equation 3.4 must be multiplied by a phase factor of \( e^{-i\varphi_{\text{NL}}} \) \{9\}.

\[
U(P) = C \int_0^{2\pi} \int_0^L E_i(\rho) e^{-ik\rho \cos(\theta - \varphi)} e^{-i\varphi_{\text{NL}}} \rho d\theta d\rho
\]

- (3.6)

Using the standard Bessel functions,

\[
J_n(x) = \frac{i^{-n} 2\pi}{2\pi} \int_0^\infty e^{i\cos \beta} e^{in\beta} d\beta
\]

- (3.7)

we get,

\[
U(P) = 2\pi CE_0 \int_0^\infty J_0(k\rho w) e^{-\left(\frac{\rho^2}{w^2}\right)} e^{-ik\rho \cos(\theta - \varphi)} e^{-i\varphi_{\text{NL}}} \rho d\rho
\]

- (3.8)

Since \( \omega \approx \alpha \), the diffraction angle; the light intensity at far field can be written as,

\[
\mathcal{I}(\alpha) = \left(\frac{2\pi}{D}\right)^2 \int_0^\infty \left| J_0(k\rho \alpha) e^{-\left(\frac{\rho^2}{w^2}\right)} e^{-ik\rho \cos(\theta - \varphi)} e^{-i\varphi_{\text{NL}}} \rho d\rho \right|^2
\]

- (3.9)

where \( D \) is the distance from the sample.

### 3.3 Numerical simulations

Considering a Gaussian beam with beam waist \( w \) and radius of curvature \( R \) propagating through a thin nonlinear film of thickness \( d \), the transmitted intensity can be described using equation 3.9 as,

\[
I(\alpha) = \left(\frac{2\pi}{D}\right)^2 \left| \int_0^\infty r dr J_0(kr \alpha) e^{-\left(\frac{r^2}{w^2}\right)} e^{-ikr \cos(\theta - \varphi)} e^{-i\varphi_{\text{NL}}} \right|^2
\]

- (3.10)

where \( k = \frac{2\pi}{\lambda}, \) \( r \) is the radial co-ordinate, \( \alpha \) is the diffraction angle and \( J_0 \) is the zeroth order Bessel function. \( \varphi_{\text{NL}} \), the nonlinear phase shift induced by a plane wave, can be described as \{2\},

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Here $\delta n$ is the refractive index variation seen by the incident wave. In the
present case however the nonlinear phase shift is also a function of the transverse
co-ordinate $r$ ($\varphi_{NL}(r)$) because of the Gaussian intensity profile, and hence we should
modify the above equation as,

$$\varphi_{NL}(r) = \frac{2\pi}{\lambda} \int \delta n(r,z) dz$$

so that we can write the Fraunhofer diffraction integral for the present case as,

$$I(\alpha) = \left( \frac{2\pi}{D} \right)^2 I_0 \int_0^\infty rdr J_0(kr) e^{-\frac{r^2}{w^2}} e^{-ik \left( \frac{r^2}{2R} + \int_0^d \delta n(r,z)dz \right)}$$

It can be seen from the above equation that the observed diffraction ring
pattern is governed by the phase factor $\varphi$, which contains two terms: the first
accounts for the wave front curvature while the second accounts for the optical
nonlinearity of the sample. Far above the onset of the nonlinear response, the latter
term dominates the phase factor and the ring pattern can be described neglecting
the wavefront curvature. In such a situation, only the SPM principal rings are
present. In contrast, just above the nonlinear response threshold both terms in the
phase factor become comparable and their interference gives rise to a smaller, fine
ring pattern at the center of the principal rings (see figure 3.4). Only one or a few
principal rings will be present for the ‘just above threshold’ condition. The small
ring pattern disappears when the sample is placed right at the focus where the
curvature is infinite (plane wave).

The nonlinear phase shift contribution is related to the intensity-dependent
refractive index of the material. If we assume a Kerr nonlinearity, the refractive
index changes can be represented as $\delta n(r,z) = n_2I(r,z)$. This change is practically
independent of the propagation coordinate (thin sample approximation), and we can write the nonlinear phaseshift term as,

$$\int_0^d \delta n(r,z)dz \approx Be^{-\left(\frac{-2r^2}{w^2}\right)}$$  \hspace{1cm} (3.14)

The quantity $B$ is defined as, $B = \Delta n \times d$, where $\Delta n$ is the maximum induced variation of the refractive index and $d$ is the thickness of the sample. Using the above equation, the Fraunhofer diffraction integral can be simplified as,

$$I(\alpha) = \left(\frac{2\pi}{D}\right)^2 I_0 \int_0^\infty rdr J_0(k\alpha r)e^{-\left(\frac{r^2}{w^2}\right)} e^{-ik\left(\frac{r^2}{2R} + B \exp\left(\frac{-2r^2}{w^2}\right)\right)}$$  \hspace{1cm} (3.15)

Figure 3.4: Photograph showing the principal SPM ring pattern and the central fine ring structure.

We did numerical simulations based on equation 3.15 for the condition of comparable phase terms, to fully characterize the SPM fine structure and its dependence on the different parameters appearing in the equation \{1\}. In the simulations, $R$ is the wavefront curvature, $w$ is the beam width and $B$ is the
maximum index variation multiplied by the cell thickness. The sign of the parameter $B$ represents the sign of the optical nonlinearity of the sample. We have used reasonable values for the parameters in the Fraunhofer integral. For instance, $B = 0.6 \, \mu m$ means that for a typical cell thickness of $60 \, \mu m$, the maximum induced variation of the refractive index is about 0.01, which is actually a reasonable value for nematic liquid crystals. The effects of beam width and curvature (magnitude and sign) and optical nonlinearity (again magnitude and sign) have been considered.

Figure 3.5 shows the simulations showing the dependence of the ring pattern on the magnitude of the radius of curvature of the beam.

Figure 3.5: Diffraction ring intensity pattern as a function of the radius of curvature ($R$) of the beam.
Figure 3.6 shows the variation of the intensity pattern with the magnitude of the nonlinearity of the sample. It can be seen that with the increase of nonlinearity, the second ring also increases in intensity. These results confirm the fact that the ring fine structure is strongly related to the magnitudes of the radius of curvature of the beam and the nonlinearity.

Figure 3.6: Diffraction ring intensity pattern as a function of the nonlinearity (B) of the sample.

The effect of the signs of radius of curvature and nonlinearity are considered in figure 3.7. Simulations show that when both have the same sign, the distribution of the fine ring structure is characterized by a central bright spot surrounded by another ring of lower intensity (figure 3.7(A) and 3.7(B)). When they have opposite signs, the distribution changes to a dark central spot surrounded by a bright thick
ring (figure 3.7(C) and 3.7(D)). These results are presented in table 3.1. Since \( R \) is fixed by the position of the cell with respect to the lens focus, and the spatial distribution of the small rings can be rapidly evaluated by the eye, this fine ring pattern structure can be used to visually identify the sign of the nonlinearity exhibited by the sample.

![Graphs showing the spatial distribution of the small low divergence rings for different signs of \( R \) and \( B \).]

"Figure 3.7: Computer simulations showing the spatial distribution of the small low divergence rings for different signs of \( R \) and \( B \). When both the beam curvature and the induced nonlinearity are either positive (A) or negative (B), the far field fine structure distribution consists of a central bright spot surrounded by a ring. If either \( B \) or \( R \) change sign, the intensity distribution changes to a dark center surrounded by a bright ring ((C) and (D))."

<table>
<thead>
<tr>
<th>( R )</th>
<th>( n_2 )</th>
<th>Fine ring spatial distribution</th>
</tr>
</thead>
<tbody>
<tr>
<td>Positive</td>
<td>Positive</td>
<td>Central bright spot</td>
</tr>
<tr>
<td>Positive</td>
<td>Negative</td>
<td>Central dark spot</td>
</tr>
<tr>
<td>Negative</td>
<td>Negative</td>
<td>Central bright spot</td>
</tr>
<tr>
<td>Negative</td>
<td>Positive</td>
<td>Central dark spot</td>
</tr>
</tbody>
</table>

"Table 3.1: Effect of the signs of beam curvature and optical nonlinearity on the fine ring pattern in the far field."
The magnitude of the nonlinearity can be found out from the number of the principal SPM rings. In the simplest situation, when the sample is at the focus, the nonlinear phase shift given by equation 3.12 can be written as,

\[ \delta \phi(r) = \delta \phi_0 e^{-\left( \frac{r}{\omega} \right)^2} \]  

which means that the wave front suffers a phase shift dependent on the transverse coordinate. Since in the focal waist of a Gaussian beam, the plane wave approximation is fulfilled, we can write the wave vector as \( k = k_z z \), while the nonlinear phase shift gives rise to a transverse component of \( k \) given by

\[ k_\perp = \frac{d(\delta \phi(r))}{dr} \]  

Because of the Gaussian shape of equation 3.16 there are two different values of \( r \), which correspond to the same slope of \( \delta \phi(r) \), that is, to the same \( k_\perp \). It means that these portions of the wave front travel in the same direction and can interfere. Therefore, the interference maxima and minima occur when

\[ \delta \phi(r_1) - \delta \phi(r_2) = m\pi \]  

where \( m \) is an integer, which is even for maxima and odd for minima.

Figure 3.8: Radial distribution of nonlinear phase shift induced on a Gaussian beam.
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The interference pattern will appear as bright and dark rings in the far field. The maximum phase shift $\delta \varphi_0$ can easily be related to the number of rings,

$$\delta \varphi_0 = \delta \varphi (0) - \delta \varphi (\infty) = N 2\pi$$  \hspace{1cm} (3.19)

or

$$N = \frac{\delta \varphi_0}{2\pi}$$  \hspace{1cm} (3.20)

Even though this is a qualitative description, it can be considered a good approximation when $N$ is large, taking into account that the experiment uncertainty is $\approx 2\pi/(N \times 2\pi) = 1/N$.

From equation 3.11, it can be seen that the maximum phase shift in a sample induced by a planar wave is given by

$$\delta \varphi_0 = \frac{2\pi}{\lambda} \delta n_0 d,$$  \hspace{1cm} (3.21)

where $d$ is the sample thickness. Using equations 3.20 and 3.21, the nonlinear refractive index can be related to the total number of rings as,

$$\delta n_0 = \frac{N \lambda}{d}.$$  \hspace{1cm} (3.22)

Hence if one knows the incident laser wavelength and the sample thickness, the nonlinear refractive index can be found by counting the principal SPM rings formed.

3.4 Experimental

An experimental test of the proposed method for the determination of the sign of the optical nonlinearity can be easily realized. For this, a highly nonlinear film is moved through the focus of a Gaussian beam from a point before the focus where the beam curvature is negative, to a point after the focus where the
curvature is positive. During this motion the changes in the distribution of the small rings pattern is carefully observed.

Three different kinds of sample have been tested, all of which are liquid crystalline materials:

Sample 1: Homeotropic cells with both surfaces treated with a solution of dimethyl octadecyl[3-(trimethoxysilyl)-propyl] ammonium chloride (DMOAP) and isopropyl alcohol. The nominal thickness of the cells, fixed by mylar spacers, is 100 \( \mu m \). Empty cells have been filled with nematic pentyl-cyanobiphenyl (5CB) in the isotropic phase and then slowly cooled down to room temperature.

Sample 2: Homeotropic doped cells with both surfaces treated with DMOAP and isopropyl alcohol. The nominal thickness of the cells, fixed by mylar spacers, is 23 \( \mu m \). Empty cells have been filled with a mixture of the nematic 5CB doped with the azo-dye, methyl-red (MR), with a weight concentration of 0.1\% in the isotropic phase and then slowly cooled down to room temperature.

Sample 3: Planar cells with only one surface treated with poly-vinyl alcohol and rubbed to obtain strong planar anchoring. The nominal thickness of the cells, fixed by mylar spacers, is again 23 \( \mu m \). Empty cells have been filled with a mixture of the nematic 5CB and MR (0.1\% by weight) following the usual procedure used for the other two kinds of sample.

The last type of cell has only one of the two glass substrates covered by the surfactant, because this procedure yields samples with a good alignment and a surface with weak anchoring, which enhance the optical nonlinear response \{10\}.

The laser source used for the measurements was a frequency doubled Nd:YVO\(_4\) CW laser with \( \lambda = 532 \) nm (Verdi, Coherent), focused by a 22 cm plano-convex lens to a diameter of \( \sim 100 \) \( \mu m \) near the sample. The laser beam falls on the sample at normal incidence, and in the case of planar cells, it irradiates the untreated surface and enters the sample as a pure e-wave. The incident laser power is of the order of 0.1 mW for the planar cells, 10 mW for the homeotropic doped cells, and 100 mW for the homeotropic undoped nematic cells. These values
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correspond to intensities of about 3 W/cm², 30 W/cm², and 300 W/cm² respectively. In planar cells a static bias (variable typically between 1.2 and 2.5 V) is applied between the substrates. The ring pattern generated is displayed on a screen 1.8 m away and is captured with a CCD camera for detailed analysis. A schematic of the experimental setup is given in figure 3.9.

![Figure 3.9: Experimental setup for recording the SPM ring patterns.](image)

3.5 Results and discussion

The low divergence rings typical of the SPM fine structure are clearly visible in all the samples analyzed, and in each case we fitted the experimental intensity distribution using equation 3.15, with \( B \) as a fitting parameter. The patterns obtained from the three different types of cell are described below in detail.

3.5.1 Undoped homeotropic cells

Santamato and Shen have reported the field-curvature effect on the diffraction ring pattern of a laser beam dressed by spatial self-phase modulation in a nematic film \([9]\). The intensity required to observe the small rings must slightly overcome the optical Fredericks threshold. The expected nonlinearity is positive since under light irradiation the refractive index changes from \( n_o \) to a higher value between \( n_o \) and \( n_e \). When the cell is before the focus (\( R < 0 \)) the distribution is characterized by a dark central spot surrounded by an intense ring, whereas when the cell is after the focus (\( R > 0 \)) the distribution changes to a central bright spot surrounded by a ring of lower intensity. According to the simulations and table 3.1, this corresponds to a positive induced optical nonlinearity. The intensity distribution fitted to equation 3.15 is shown in figure 3.10. The fitting parameter \( B \) is
positive, as expected. The value of $B$ found by the fitting procedure corresponds to a maximum induced birefringence ($\Delta n$) of about 0.007 and to a nonlinear coefficient ($n_2$) of the order of $10^{-5}$ cm$^2$/W, which is typical of the giant optical nonlinearity of liquid crystals responsible for SPM in this kind of cell \cite{2}.

![Figure 3.10: Experimental far field intensity distribution of the small rings in the case of undoped homeotropic cells. The line represents the fit based on equation 3.15. The cell lies before the beam focus (negative beam curvature) in (A), and after the focus (positive curvature) in (B).](image)

3.5.2 Homeotropic cells doped with methyl red

In this case the presence of the absorbing dye produces, at the intensity used, a nonlinear response of thermal nature. The optical nonlinearity associated to this kind of response is expected to be positive because $dn_0/dT$ is a growing function of $T$; that is, the ordinary refractive index increases with the sample temperature. Since the incident Gaussian beam has a maximum intensity at its centre, the cell temperature will be higher in this region and lower in the beam wings, with the refractive index following the same trend.

The spatial distribution of the low divergence rings is shown in figure 3.11 for both positions of the cell with respect to the lens focus. Again, negative
curvature corresponds to a dark central spot surrounded by an intense ring, and positive curvature corresponds to a central bright spot surrounded by a ring of lower intensity, which is in agreement with an induced nonlinearity of positive sign. Also, the sign of $B$ derived by fitting the intensity curve with equation 3.15 is positive, as expected.

![Figure 3.11](image)

**Figure 3.11:** Spatial distribution of the fine structure rings in the case of doped homeotropic cells. Negative curvature corresponds to a dark central spot surrounded by an intense ring, and positive curvature corresponds to a central bright spot surrounded by a ring of lower intensity. The induced optical nonlinearity is positive and has a thermal origin.

### 3.5.3 Planar cells doped with methyl red

In the case of planar cells, both SPM principal rings and fine structure can be obtained only when an external bias is switched on and is kept lower than the Fredericks threshold voltage \{11\}. The behaviour of the spatial distribution of the low divergence rings is different with respect to that of homeotropic samples. This time, when the cell is before the focus ($R < 0$), the distribution is characterized by a central bright spot surrounded by a ring of lower intensity, whereas if the cell moves beyond the focus ($R > 0$) the distribution changes to a dark central spot surrounded by an intense ring. According to the simulations, this corresponds to a negative induced optical nonlinearity. The occurrence of a negative nonlinear response in planar cells is in agreement with the phenomenological model proposed by Lucchetti et.al. \{11\}. The model states that the incident light modulates the effective internal voltage acting on the surface charge density of ions in such a way that the external bias produces a director reorientation towards the homeotropic
configuration only, corresponding to the centre of the incident Gaussian beam. In this way, the refractive index seen by the incident light is \( n_e \) in the beam wings, and has a value between \( n_o \) and \( n_e \) at the beam centre, which gives rise to self-phase modulation due to a negative nonlinearity. Figure 3.12 shows the distribution of the intensity transmitted by the cells in the two positions: before (figure 3.12(A)) and after (figure 3.12(B)) the lens focus. Note that these are reversed with respect to the curves shown in figure 3.10. The fitting parameter \( B \) is negative, as expected. Its value is -0.8 \( \mu m \) in one case and -0.7 \( \mu m \) in the other. This corresponds to a maximum induced birefringence (\( \Delta n \)) of about 0.03, which means a nonlinear coefficient \( n_2 \) of the order of \( 10^{-2} \) \( cm^2/W \), consistent with the recently observed nonlinear response of planar cells of 5CB doped with methyl red \{11\}.

**Figure 3.12:** Experimentally determined far field intensity distribution of the small rings in the case of a planar cell of 5CB doped with MR placed before (A) and after (B) the lens focus. Solid lines represent theoretical fits with equation 3.15. In this case the induced optical nonlinearity is negative.

### 3.6 Conclusions

In conclusion, we have devised a fast and easy method to determine at a glance the sign of the optical nonlinearity of highly nonlinear film samples. The
technique is similar to the conventional z-scan \{12\} except for the fact that no aperture is necessary to determine the sign of the nonlinearity. Since the number of SPM principal rings is directly related to the value of the induced birefringence, the method proposed allows a simple evaluation of both the sign (by means of the SPM fine structure) and the value (by counting the SPM principal rings) of $n_z$. The method is independent of the type of nonlinearity that causes the ring pattern, as demonstrated by measurements in different types of nematic cell, and is expected to work for every kind of highly nonlinear medium.
References


