POLARISATION SOLITONS IN PHOTOREFRACTIVE CRYSTALS WITH STRONG OPTICAL ACTIVITY: THE VLAD'S DISCOVER

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Abstract: This paper describes the results achieved by Valentin Ionel Vlad in the field of soliton formation in photorefractive crystals with high optical activity. Both through an accurate theoretical and experimental study, it has been shown that even in these materials the soliton regime can be reached thanks to a very high phase self-modulation of the polarisations involved, which push the field to rotate the polarisation plane much faster. This nonlinear acceleration initially generates beams with different transversal polarisation orientations: however, once the soliton regime is reached, the polarisation is homogenized along the whole transversal direction of the beam, rotating according to levels of elliptical polarisation interconnected through a limit trajectory of the state on the Poincaré sphere.

Keywords: spatial soliton, polarisation soliton, photorefractive soliton, two-photon soliton, photorefractivity, nonlinear optics, Valentin I. Vlad

Introduction

This work is the result of the calm but indomitable enthusiasm that Valentin I. Vlad had towards research and life in general, which with his constant presence he instilled in me, his and my collaborators.

This work begins in the spring of 1997 when Valentin Vlad came as visiting scientist of the ICTP (International Centre of Theoretical Physics) in Rome to my laboratory. At that time I was working on the formation of Kerr-type spatial solitons in planar glass guides [1-2]. When he arrived he proposed me to work on photorefractive materials and, in particular, to study the soliton formation in Bismuth Silicone Oxide (BSO - $Bi_{12}SiO_{20}$) crystals. I didn't know what photorefractivity was: but that encounter literally changed my life!

Spatial solitons

A soliton beam is a light beam that does not diffract because the light is able to modify the refractive index of the surrounding material, creating a kind of channel within which the light is self-confined, propagating as if inside a waveguide or a fibre optics. There are many nonlinear processes that can vary the refractive index. The first and simplest known effect is the optical Kerr effect; it is associated with a third order nonlinearity of the type:

$$\varepsilon = \varepsilon_0(1+\chi) = \varepsilon_0\left(1+\chi^{(1)}+\alpha\chi^{(3)}I\right) = \varepsilon_0(\varepsilon_{r-linear}+\epsilon I) \qquad (1)$$

which leads to a refractive index linearly proportional to the luminous intensity I:

$$n = n_0 + n_2 I\,. \qquad (2)$$

The first theoretical study on the formation of self-confined beams using optical Kerr nonlinearity dates back to 1964 [3] by E.T. Chiao, E. Garmire and C.T. Townes who, in that year, received the Nobel Prize for his studies on lasers and masers. In the paper the authors

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interpreted this phenomenon as follows: "*We shall discuss here conditions under which an electromagnetic beam can produce its own dielectric waveguide and propagate without spreading*". The complete analytical theory of the existence of spatially self-confined beams was published only 8 years later, in 1972, by V. E. Zakharov and A. B. Shabat [4] while the first experimental proof arrived only 13 years later, in 1985 by the hand of A. Barthelemy, S. Maneuf and C. Froehly [5]. In this paper they observed the selfocusing and self-confinement of a laser beam that propagated inside a cell containing liquid CS_2. Subsequently, in 1990, Aitchinson et alii [6] succeeded in obtaining solitons within a solid material, in a planar glass guide. This experiment on the one hand showed that the soliton beams could be used in chips and that they could be exploited for important applications, on the other hand it showed a big limit of the optical Kerr nonlinearity due to its very low value, being a nonlinearity of the 3° order. Very high light intensities must be used to observe important effects, as high as gigawatts per square centimetre. Such high intensities can be achievable with ultrashort laser pulses, as short as femto- or picosecond, but nevertheless they are hardly exploitable in possible technological applications. It was necessary to break down this barrier by using a technology that could work at much lower optical fluences.
The answer to this problem arrived 2 years later, in 1992, when Segev et alii [7]demonstrated experimentally, and in the following year theoretically too [8], that by exploiting the photorefractive nonlinearity it was possible to obtain selfconfined laser beams.
Photorefractivity is a second-order nonlinear process as it exploits the static Kerr effect also called the Pockels effect or electro-optical effect, according to which it is possible to change the refractive index of a crystal by applying a static field. Its advantage over optical Kerr is that being an inferior order nonlinear it requires less intense fields, shifting the excitation of the nonlinear intensity of the light to a static field, much easier to generalize. With photorefractivity, the light intensities dropped considerably, reaching values of the milliwatt per square centimeter, about 10-12 orders of magnitude lower! The technique of photorefractivity generates a static electric field by locally exciting a charge distribution (Coulomb field), for example by carrying out electronic transitions starting from trap states due to lattice defects, impurities or suitable doping. Due to the transitions, the electrons are transferred into conduction states and thus can migrate freely in the crystal while the holes remain locked in their local positions. Using laser beams with a transverse bell-like profile (typically Gaussian beams), charge distributions and therefore static fields with bell-like profiles can be generated, which consequently produce bell-shaped variations in the refractive index.
A problem arises here: the electro-optical effect produces a decrease in the refractive index and not an increase, which instead is necessary to create a guiding optical structure. This is a direct consequence of the physics of optical crystals. A second order nonlinearity is, by its nature, anisotropic and therefore can only be observed in crystals. The refractive index of a crystal is represented by the ellipsoid of the indices:

$$\frac{x^2}{{n_x}^2}+\frac{y^2}{{n_y}^2}+\frac{z^2}{{n_z}^2}=1. \quad (3)$$

Therefore, the refractive index variation is described by variational terms such as:

$$\Delta\left(\frac{1}{n_i^2}\right)=\sum_j r_{ij}E_j(0) \qquad (4)$$

where r_{ij} are the electro-optical coefficients. Equation (4) corresponds to writing a nonlinear refractive index of the type:

$$n_i[E(0)] = n_{i,0} - \frac{1}{2}\sum_j n_{i0}^3\, r_{ij} E_j(0). \quad (5)$$

As you can clearly see, the electro-optical effect decreases the refractive indices. Due to this decrease, mainly dark solitons were observed, that is, the diffraction-free propagation of shadows within a luminous background [9-10]. To create bright solitons it is necessary to exploit a small "trick": a static and uniform bias field is applied to the photorefractive crystal which, by exploiting the electro-optical effect, decreases the refractive index of the whole material. The light absorption generates two distinct populations of charges, one electronic in nature, that migrates due to the effect of the bias field, and one of holes, that remain put into their physical places of excitement. The separation of these two distributions produces an induced field capable of screening the applied field, similar to what happens in a metal due to the Faraday effect. Consequently, in the direction of the bias (which usually coincides with the optical axis of the crystal) the local electric field decreases or completely deletes, bringing the refractive index back to its linear unmodified value. By doing so, that is, by exploiting the screening of the bias field, it is possible to generate a local variation of the refractive index greater than the contour, within which the light can selfconfine, generating bright solitons.
From this point begins Valentin Vlad's adventure and discoveries: as previously mentioned, the crystal anisotropy requires a fine control of the directions of application of the bias and of the polarisation of light that must coincide. A polarisation along the crystallographic direction x will feel the n_x index, a polarisation along y will feel n_y and a polarisation along z will feel n_z. By applying the bias field for example along x, there will be a variation in the index n_x .... and not of others. Therefore, only the light polarisation parallel to the bias experiences theoptical nonlinearity; an orthogonal polarisation will not feel the nonlinearity, following a linear diffraction due to a constant refractive index.
The scientific challenge that V. Vlad came to me in the spring of 1997 was to observe solitons in a material with a high optical activity. Optical activity is that physical phenomenon whereby light, during propagation, rotates the direction of polarisation. This obviously was in clear contradiction with the basis of photorefractivity just explained: *by rotating the polarisation plane, the light will no longer feel the nonlinearity when its polarisation arrives orthogonal to the direction of bias application. Therefore, during propagation, light should pass from nonlinear selfocusing to linear diffraction conditions and, consequently, the soliton regime might not be achieved.*

Polarisation solitons

We began to investigate the problem which soon appeared very complex. We decided to follow 3 parallel paths: the first occurred in Bucharest where Valentin Vlad and Vasile Babin, a very smart and prepared theoretician, tried to find analytical solutions ; at the same time in Rome an experimental measuring apparatus was set up to observe (1+1)D solitons in BSO crystals; contemporarily, a numerical computation algorithm was realised to simulate the nonlinear propagation of light using an FDTD code (Finite Differences in the Time Domain).
It was a very hard job and it took us 3 years to get the first results.
Vasile Babin developed an approximated analytical solution of the nonlinear problem that was initially published in the Proceedings of the Romanian Academy [11] and later, in the full version, in Physical Review E [12]. His work started from the following set of nonlinear Helmholtz equations [12]:

$$\vec{\nabla}^2\vec{E}-\frac{1}{c^2}\left[\vec{\varepsilon}\times\frac{\partial^2\vec{E}}{\partial t^2}+\frac{g}{k}\left(\vec{\nabla}\times\frac{\partial^2\vec{E}}{\partial t^2}\right)\right]-\frac{n_0}{c}\alpha\frac{\partial\vec{E}}{\partial t}=0 \qquad (6)$$

where $\vec{\varepsilon}$ is the nonlinear tensor of the dielectric constant, g is the gyration constant that describes the optical activity and α the linear absorption. Neglecting absorption and performing some change of coordinates, equation (6) can be rewritten with the explicit notation:

$$\nabla_\perp^2\begin{bmatrix}A_x\\A_y\end{bmatrix}+\begin{bmatrix}(\varepsilon_{xx}-1) & (\varepsilon_{xy}+ig)\\(\varepsilon_{yx}-ig) & (\varepsilon_{yy}-1)\end{bmatrix}\begin{bmatrix}A_x\\A_z\end{bmatrix}=0 \quad (7)$$

where the diagonal coefficients ε_{ii} =1 for cubic crystals (like BSO) while the nondiagonal ones

$$\varepsilon_{ij}=\frac{n_{0i}^2 r_{ij}\mathrm{E}_{bias-i}}{1+\frac{I_i+I_j}{I_{background}}} \qquad (8)$$

describe the optical nonlinearity. Note that this expression contains both the static bias and the total light intensity normalized to the background one. Eq. (8) shows a saturable dielectric constant, and consequently a saturable refractive index too, fundamental condition for obtaining stable solitons in 2 dimensions (the Kerr-optical type solitons are not stable in 2D but only in 1D precisely due to the lack of nonlinear index saturation).

The resolution of equation (7) is quite complex: the interested reader can consult the ref. [12] for the complete calculation. Here I would like to point out that all the obtained solutions lead to pulsating solitonic behaviors (as for example in fig.1 - [12]) as numerically and experimentally observed.

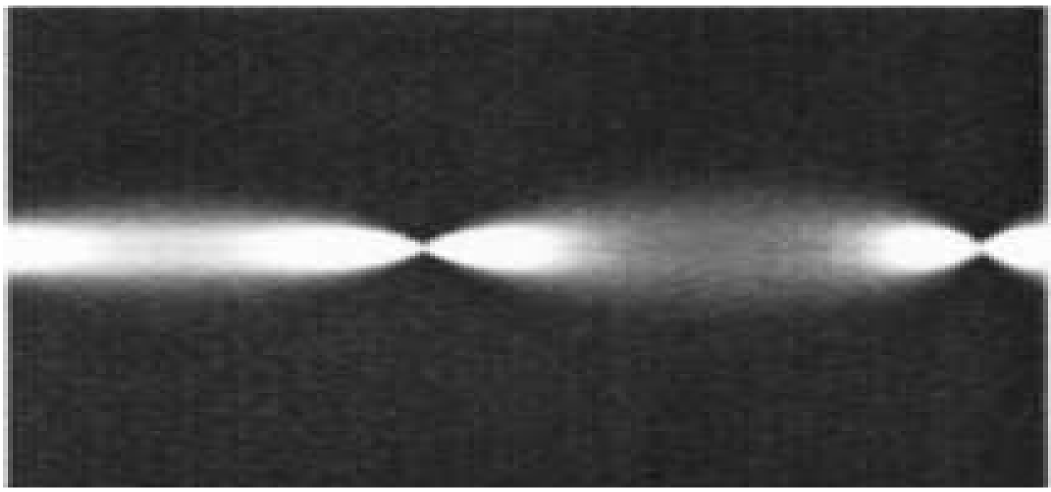

Fig. 1 The approximate analytical solutions of the nonlinear equations for materials with high optical activity lead to pulsing self-confined beams [12].

On the same time, the numerical simulations gave their first results, showing that the solitonic regime was reachable at very high biases [13], much higher than expected.

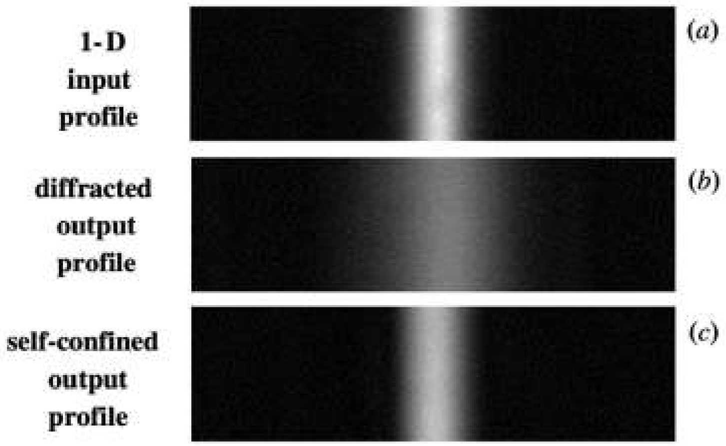

Fig.2 Experimental images of a 1D light beam at the input and output planes of the nonlinear crystal. The linear diffraction shows a large beam which results selfconfined as soon as the electric bias is applied to the crystal.

In fact, the achievement of soliton regime was achieved both numerically and experimentally by applying extremely intense biases, higher than those found in the literature: in 1996 Wieslaw Krolikowski et alii [14] reported that it is possible to achieve selfocusing in BSO at bias fields between 1 and 10 kV/cm. Similar results were reported for other materials such as in BTO [15]. Our work [13] demonstrated that the bias fields must be raised up to 30-40-50 kV/cm in order to reach effectively the solitonic confinement. At that moment, we did not understand why was such high bias field needed to selfconfine light: the explanation came later, when we pushed the experimental study to the (2+1)D regime.
In 2003 we published on Physical Review E [16] the first experimental observation of a two-dimensional soliton in BSO. This paper was not so much important for the effective demonstration of the 2D confinement, but because it introduced for the first time the study of the polarisation state of the soliton. Let’s see which information we derived.
The numerical model for light propagation that we solved was based on the following coupled equations, where only the y electric field component experiences the nonlinearity:

$$\begin{cases} \left(2\mathrm{ik}\frac{\partial}{\partial \mathrm{z}} + \nabla_\perp^2\right)\mathrm{E_x} - \mathrm{igE_y} = 0 \\ \left(2\mathrm{ik}\frac{\partial}{\partial \mathrm{z}} + \nabla_\perp^2 + \delta\varepsilon_{\mathrm{NL}}\right)\mathrm{E_y} + \mathrm{igE_x} = 0 \end{cases} \qquad (9)$$

where $\delta\varepsilon_{\mathrm{NL}}$ describes the saturating photorefractive nonlinear dielectric constant:

$$\delta\varepsilon_{\mathrm{NL}} = \frac{-kn_0^2 r_{41}\mathrm{E}_{bias}}{1 + \frac{I_{soliton}}{I_{background}}} \qquad (10)$$

As output of the numerical simulations, we plotted both the distribution of light intensity during propagation (fig. 3) and the polarisation state of the light (fig. 4). The intensity distributions (fig. 3) show an almost complete selfconfinement at biases above 35 kv/cm. This was a very important input for the experiments, which fixed the testing conditions to be reached. The analysis of the polarisation state showed that the light assumed different polarisation orientations along the beam transverse profile (fig. 4). This was an absolute novelty, that was also experimentally verified by monitoring the 2 crossed output polarisations, as shown in figure 5.

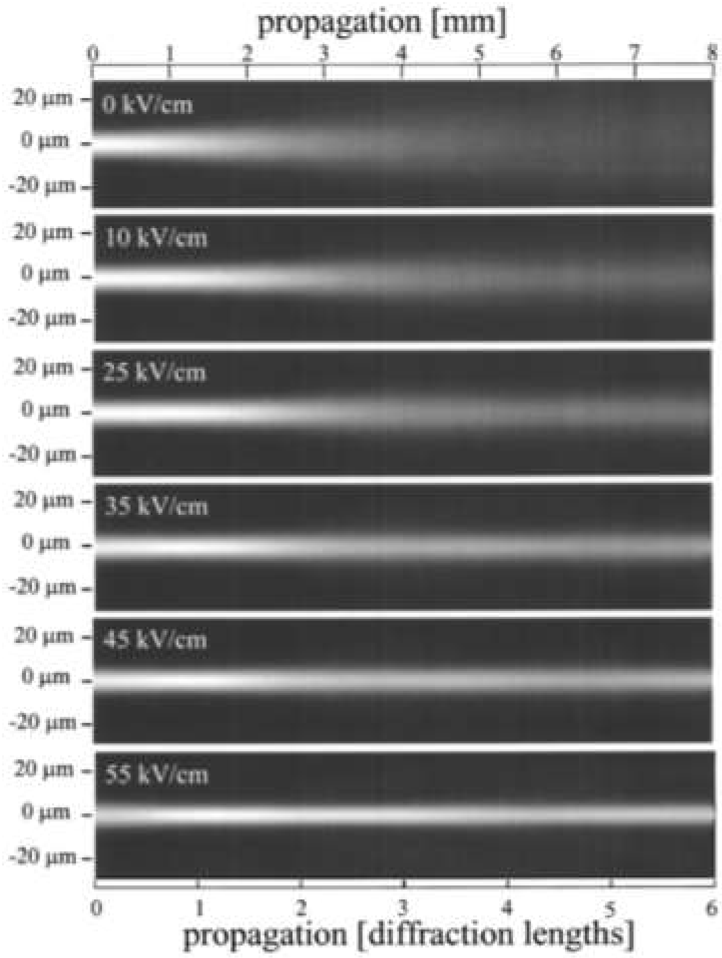

Fig.3 Evolution of the light beam propagation for different bias fields [16].

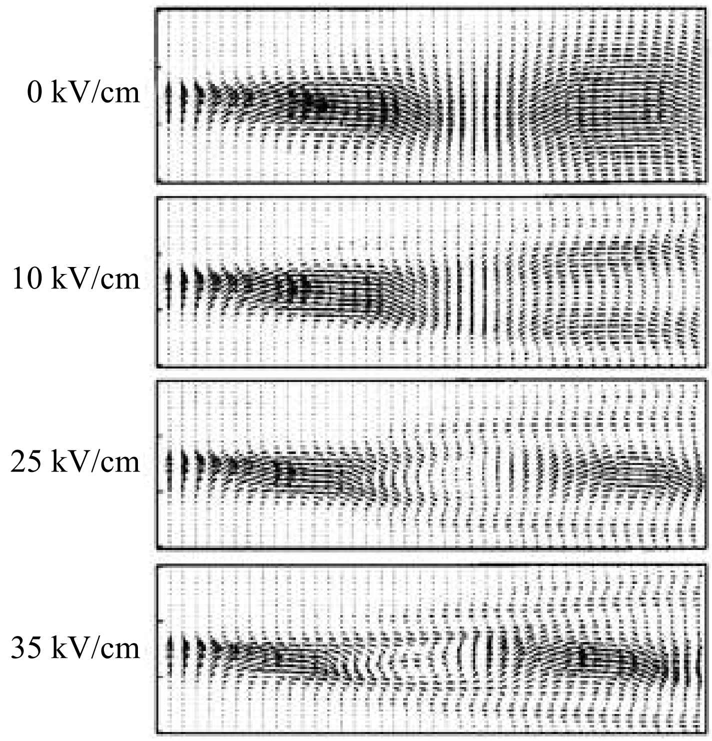

Fig.4 Evolution of the light polarisation for different bias fields [16].

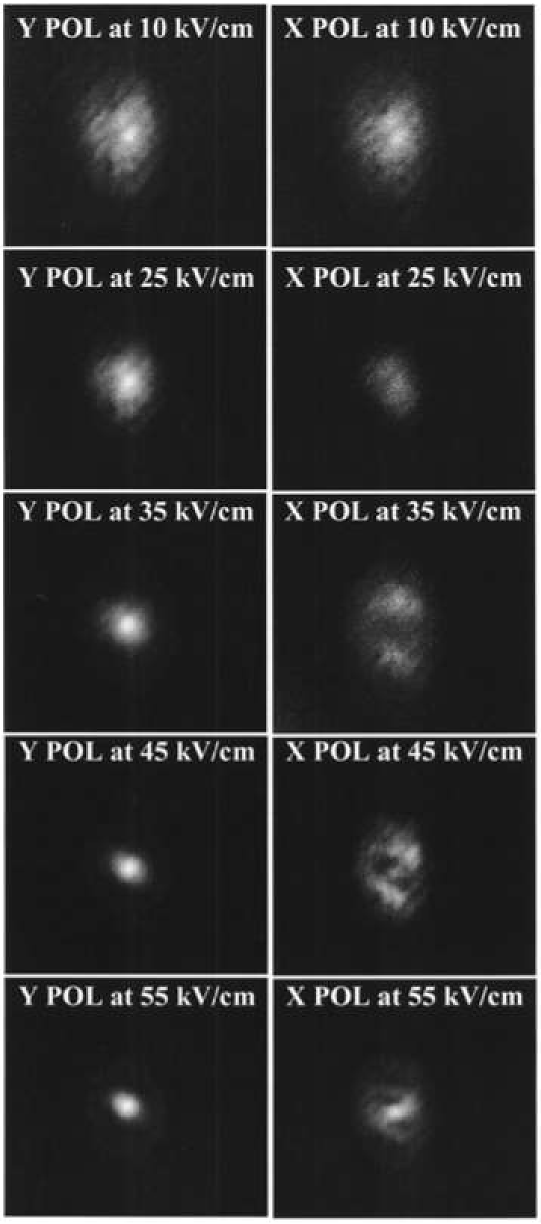

Fig.5 Experimental images of the two cross-polarisation intensities as function of the applied bias [16].

Initially, a light beam with linear polarisation at 45° with respect to the two transverse X and Y directions was injected. By applying a bias field at 25 kV/cm, we observed that the X polarisation was depleted, a sign of a nonlinear rotation of light polarisation. At 35 kV/cm this nonlinear rotation gave rise to non-uniform transverse light distributions: the core of the beam was in the Y polarisation while the edges remained X-polarised. At 45 kV/cm a further acceleration of the rotation of the polarisation forced the centre of the beam to return to the X-polarisation. For this bias the X-pol was constituted of 3 different parts: the beam edges and core were bright while the intermediate zone was dark. Reaching 55 kV/cm, the X-polarisation homogenized. For this bias, the 2 orthogonal polarisations had almost equal intensities.
It was particularly enlightening to study the polarisation evolution on the Poicaré sphere, reported both in the paper [16] and, later on, in [17] where a complete study of the soliton polarisation behavior was reported. Here, we reported the existence curves of polarisation solitons as a function of the input intensity, of the applied bias field and of the different input polarisation states. The propagation dynamics plotted on the Poincaré sphere (fig. 6) showed

that according to the applied bias field, the light no longer remains linearly polarised but becomes elliptical, evolving during the during the establishment of the soliton regime, until a limiting trajectory is reached, characteristic of polarisation-soliton state.

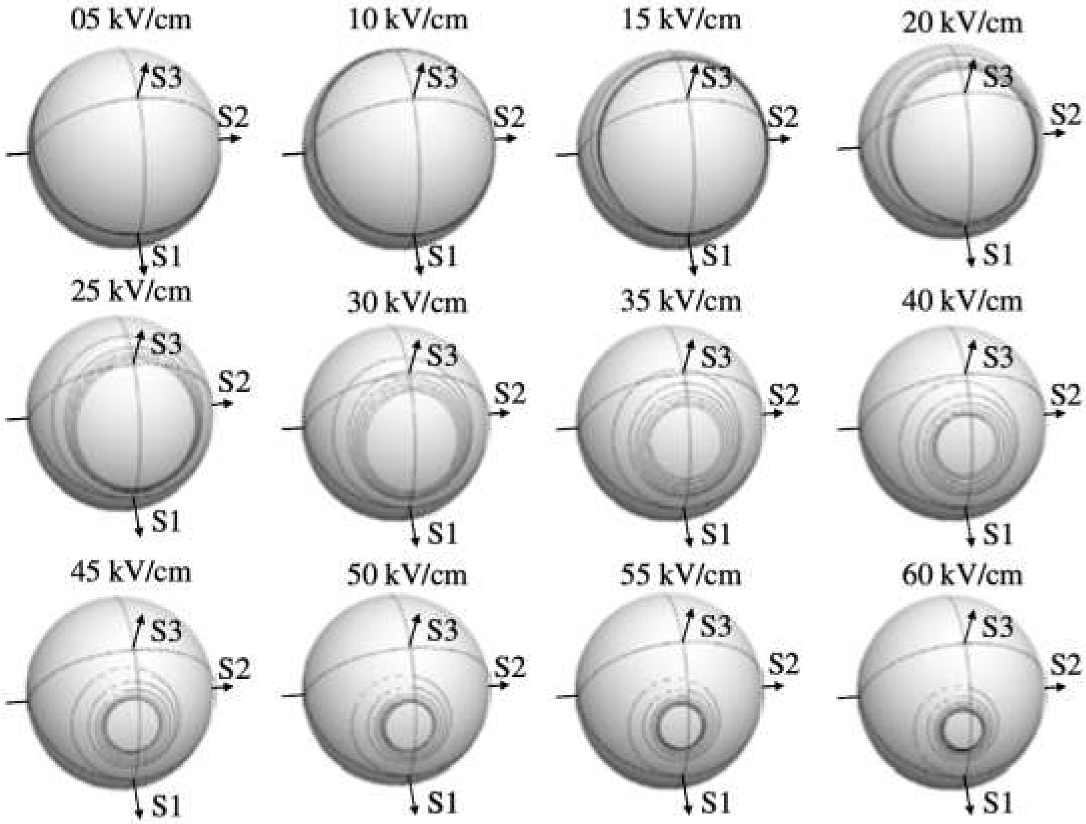

Fig.6 Plots of the polarisation evolution on the Poincaré sphere [17] as function of applied bias fields. Solitonic regimes correspond to the limit almost-circular trajectories.

This phenomenon is due to the phase modulation that each light polarisation experiences because of the optical nonlinearity, which induces a nonlinear polarisation rotation. Light moves into a highly nonlinear state for which its electric field stabilizes in a series of elliptical polarisation states interconnected through the limit trajectory.

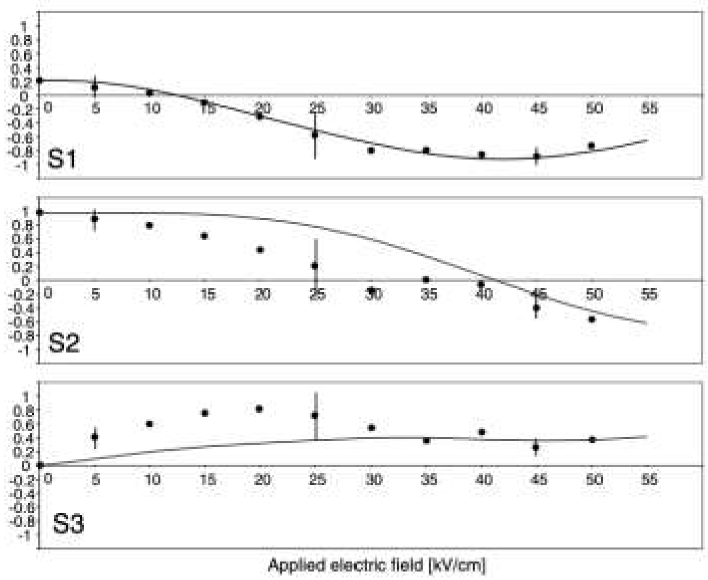

Fig.7 comparison of the experimental output polarisation states (points) with the simulations (lines) [17]

These simulations have been experimentally verified, comparing the numerical results of the polarisation leaving the crystal with the experimental ones: in fig. 7 the experimental (points) and theoretical trends (lines) of the 3 Poincaré coordinates coming out of the crystal are reported as a function of the applied bias field. The observable discrepancies for the S2 and S3 coordinates at low and medium bias fields are due to the difficulty of identifying a single polarisation state at these regimes since, as previously shown in fig. 4, the light beam showed transversally different polarisation orientations at these regimes.

Two-colour solitons

These studies also led to the first finding, both theoretically and experimentally, of photorefractive solitons that exploited both 2-photon and 2-step absorption processes [18]. This was an extremely hot topic of great interest: however, we probably got the title of the article wrong, which did not have the due echo: in fact, 2 years later in 2005, Chunfeng Hou et alii published the article "*Spatial solitons in two- photon photorefractive media*"[19] analogous using exactly our same model but without referencing our work.
This study originated from the observation that it was possible to reach the soliton regime at wavelengths that were not naturally absorbed by the material (in this case red light at 633 nm) and that therefore could not originate by themselves the space charge field necessary for inducing the screening of the bias field. For the explanation of this process we considered that the application of a uniform illumination (in this case in the green) of the sample could induce two-step transitions: the uniform green illumination was absorbed, bringing the electrons in an intermediate state from which a second red beam transferred them to the conduction band. The screening field got the transverse profile of this last red beam and consequently induced its selfocusing. This two-step process originated a slightly different nonlinearity from that for single-step absorptions such as [18]:

$$\delta\varepsilon_{\mathrm{NL}} = -kn_0^2 r_{41}\mathrm{E}_{bias}\frac{1 + RI_{soliton}}{1 + \eta RI_{soliton}} \qquad (11)$$

where R and η are suitable nonlinear coefficients.
By exploiting the nonlinear variation (11), it is possible to obtain soliton confinement observable also experimentally (fig. 8).

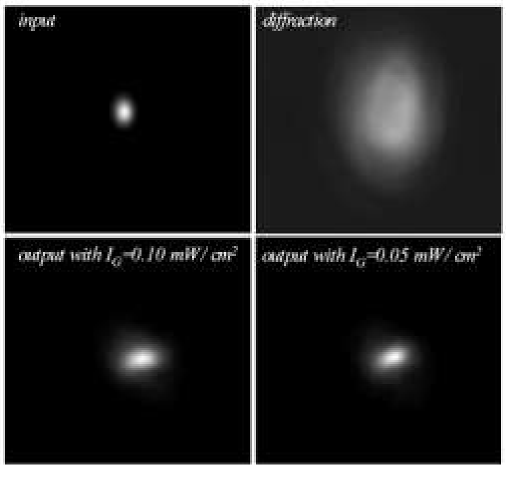

Fig.8 Formation of solitons using a two-step absorption process.

Conclusions

Experimental work on soliton formation in crystals with high optical activity has been absolutely pioneering, showing for the first time that in these materials, optical solitons are

associated with specific soliton states of polarisation. A self-confined beam is identified by a specific trajectory of its polarisation, limited in a boundary trajectory defined and well identified in the Poincaré space.
The Romanian Academy of Sciences, in December 2004, awarded Valentin Vlad, Vasile Babin, Mario Bertolotti and myself the Dragomir Hurmuzescu prize for our studies on the "formation of spatial solitons in photorefractive media".
The continuous and tireless dedication to work, the calmness of the ways but the resolute determination of Valentin Vlad, as well as his scientific ability to tackle a problem under different aspects have allowed all this to happen. Valentin Ionel Vlad was a great scientist who pushed scientific knowledge beyond the known limits thanks to his studies.

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