Nearly-forbidden optical Transitions, Nanooptics, opals, ...

Daniel Bloch
Laboratoire de physique des lasers, UMR7538 du CNRS et de l'Université Paris13
99 av JB Clément, F-93430 Villetaneuse
daniel.bloch@univ-paris13.fr

The team "Atomic Spectroscopy at Interfaces" (SAI) of LPL has mostly an experimental activity, with optical techniques sensitive to depths smaller than the optical wavelength, aiming at the measurement of long-range surface interactions (Casimir-Polder interaction, which generalizes the known "van der Waals interaction", limited to the near-field regime). More marginally, in purely theoretical approaches, or through co-operations, we have been interested into problems connected to chirality.

Theoretically, we have considered the interaction of an optical beam bearing orbital angular momentum (OAM) with nearly-forbidden transitions. This means going beyond the dipolar approximation common in optics because optical wavelengths are much larger than the size of the atom-like "particles" which are used as the elementary components of a photo-detector. One of the major goal was the understanding of a process where a beam, such as a Laguerre-Gaussian beam, susceptible to carry several units of orbital momentum per photon, could exchange at an elementary level (*i.e.* one "quantum") with matter. In most exchanges, the angular momentum is distributed between external and internal angular momentum. The light intensity of these beams bearing orbital momentum, such as the Laguerre-Gaussian beams, is null on-axis. Nevertheless, a transfer between the light beam and matter can occur on the nearly forbidden quadrupole electric transition (E2), as a consequence of electric field gradients on the axis of the beam. At centre, matter absorbs one photon bearing a $2\hbar$ orbital momentum, in a transfer limited to internal degrees of freedom [1]. Practically, such effects are to be considered, only for a strongly focused beam, beyond the paraxial approximation (focusing typically on one wavelength, *i.e.* in a nano-optics regime). This regime allows to consider that a chirality is associated, on a local basis, to the topological charge of the beam. This strongly differs with the macroscopic situation and for regular electric dipole transitions (E1), when despite the apparent chirality of OAM bearing beams, no selectivity appears in the interaction with chiral matter [2].

In experiments performed by the group of T. Yatsui at Tokyo [3], soft nanoetching on ZrO_2 nanostructures is obtained by a long duration, low-power, 325 nm irradiation under the normal incidence. The interface with the nanostructure induces locally a spatially-structured light-field, which makes efficient the (nearly-forbidden) magnetic dipole transition (M1). This transition allows locally to dissociate ambient molecular oxygen, yielding a spatially selective etching. A specific proof for this interpretation [3] is the dependence of the nano-etched region, relatively to the incident polarisation (parallel, or perpendicular to the initial nano-grating). A more complex irradiation (oblique incidence, or spatially structured beam, or in a plane with a specific orientation relatively to the direction of the initial nano-grating), and polarisation-structured beam, could break the initial symmetries, perhaps up to inducing modes with a chiral structure As shown for E2 transitions, the efficiency of M1 transitions is directly related to the spatial derivatives of the electric field, and a well-chosen microscopic geometry at the edge of the sample could allow to define modes with a specific chirality.

We have also studied the spectroscopy of an atomic gas confined in the interstitial regions of an opal (a self-organized arrangement of glass nanospheres). In a preliminary study, we could notice that a single layer of organized mono-disperse spheres exhibits birefringence properties [4]. The intensities of the diffracted beams, distributed along the summits of an hexagon, depend, for each pair of diffracted beams, on the direction of the incident (linear) polarisation relatively to the direction of the opal self-organization. An irradiation more complex than a simple plane wave in linear polarisation, or an on-purpose nanostructure, more complex than the one produced by the organization of nanospheres, may induce optical fields with specific chiral properties

[1] V.V. Klimov, D. Bloch, M. Ducloy, J.R. Rios Leite, "*Mapping of focused Laguerre-Gauss beams: The interplay between spin and orbital angular momentum and its dependence on detector characteristics*" Phys Rev A **85**, 055384 (2012)
[2] F. Araoka, T. Verbiest, K. Clays, and A. Persoons, " *Interactions of twisted light with chiral molecules: An experimental investigation*" Phys Rev A **71**, 055401 (2005)
[3] T. Yatsui, T. Tsuboi, M. Yamaguchi, K. Nobusada, S. Tojo, F.Stehlin, O.Soppera, D. Bloch, "*Optically controlled magnetic-field etching on the nano-scale*", Light: Science and Applications **5**, e16054 (2016)
[4] E. Moufarej, I.Maurin, I. Zabkov, A. Laliotis, P. Ballin, V. Klimov, D. Bloch, "*Infiltrating a thin or single layer opal with an atomic vapour: sub-Doppler signals and crystal optics*", EPL **108**, 17008 (2014)

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Daniel BLOCH

Laboratoire de Physique des Lasers

Université Paris13 *(Sorbonne Paris-Cité)* ***and CNRS****, Villetaneuse*

Outline

Team at LPL "Atomic spectroscopy at interfaces"

Casimir- Polder interaction, surface polariton modes, thermal emission in near-field ... ($\rightarrow$ **structuring of surfaces** : **opal** and more)

****

With Brazil (nonlinear optics -exp^{tal}) + Russia (theory)

Laguerer-Gauss beam and *(very)* **strong focusing**

B, ∇E "forbidden" transitions, chiral selectivity

At the origin of our theoretical interest

Linear spectroscopy: to elucidate $l\hbar$ exchange **1photon/1atom**

NLO Tabosa experiments (Recife, Brasil)

on resonant atoms in the late 90's**:** (cold) atom**s** and photon**s**

*****

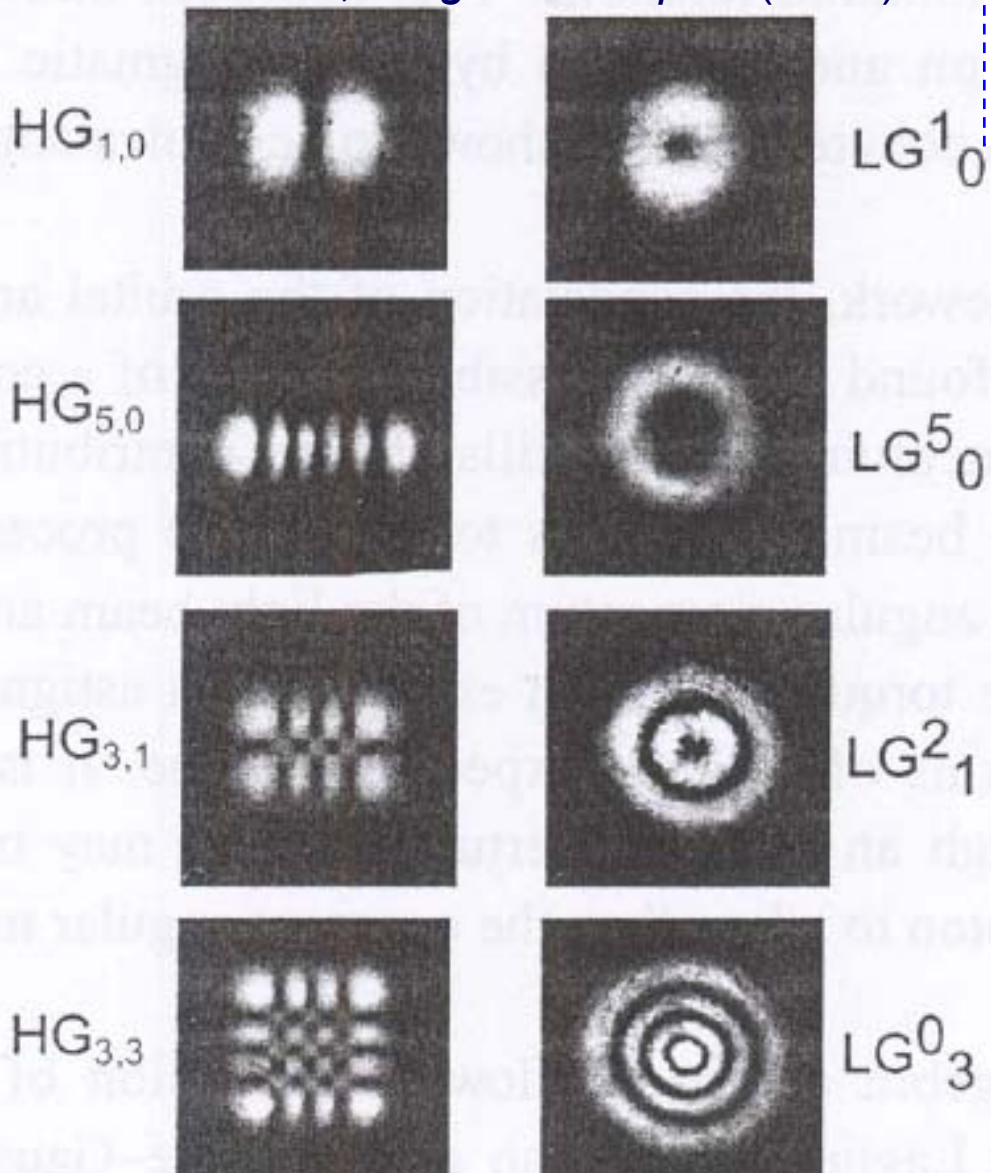
In Japan

"M1" transitions of O_2 for local dissociation at a nanostructure

a plane wave irradiation becomes locally structured

Hermite-Gauss and Laguerre-Gauss

in Allen *et al.* , *Progress in Optics*(1999)



LG beam
observed
in "intensity"

Laser field as a Gaussian beam

$$u(r, \varphi, z) = \exp -[r^2/w^2(z)]\, LG_{p,l}[r/w(z)] \cdot \exp il\varphi \exp -i[kz + \Phi(r,z)]$$

LG beam carries
Orbital **A**ngular
Momentum (1992)
$l\,\hbar$

Singular optics

V Klimov et al.

Maxwell eqs. are vectorial

→ no electromagnetic field

exhibits axial symmetry

Assuming an axial symmetry for E with phase factor exp $il\varphi$

$$\mathbf{E} = \{E_x(r,z), E_y(r,z), E_z(r,z)\}e^{il\varphi}$$

One derives for $\underline{B}$ from Faraday $ik\underline{B} = rot\ \underline{E}$:

$$B_x = -\frac{i}{k}\left(\frac{\partial E_z(r,z)}{\partial r}\sin\varphi + \frac{E_z(r,z)}{r}il\cos\varphi - \frac{\partial E_y(r,z)}{\partial z}\right) \times e^{il\varphi} \neq B_x(r,z)e^{il\varphi}$$

Not the whole vectorial structure exhibits the axial symmetry $e^{il\varphi}$

maximal axial symmetry $\mathbf{E} = \{E_x(r,z), E_y(r,z), E_z(r,z,\varphi)\}e^{il\varphi}$

LG or Bessel defined transversally → longitudinal component

→ **<u>E</u> and <u>B</u> structures are NOT ANALOGOUS !** (***nano-optics !***)

Beyond the PARAXIAL approximation

$$\mathbf{E} = E_0 \frac{w_0}{k} \left\{ k\alpha U, k\beta U, i\left(\alpha \frac{\partial U}{\partial x} + \beta \frac{\partial U}{\partial y} \right) \right\} e^{ikz}$$

longitudinal component $\nabla E = 0$

U is for the transverse shape (standard LG)

Faraday

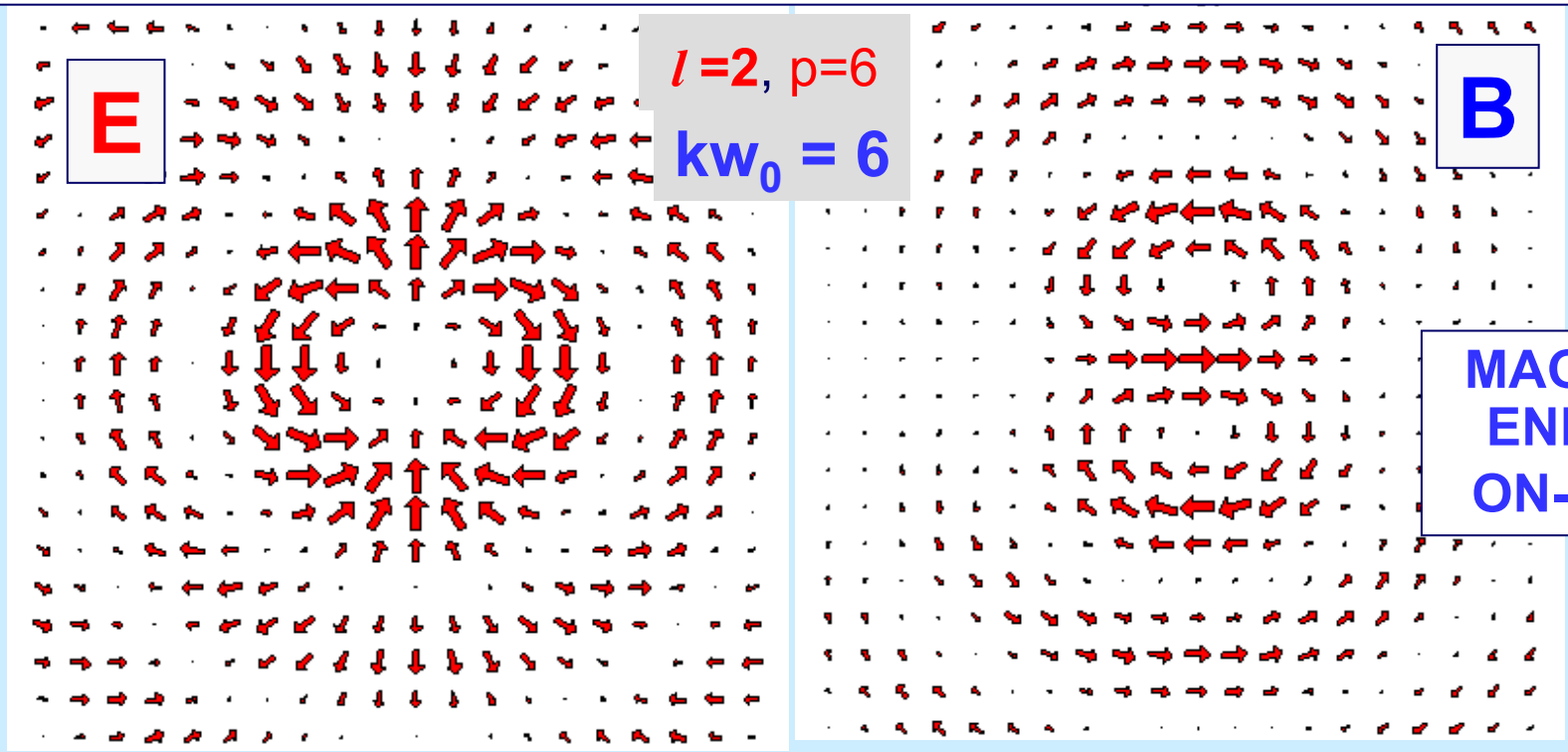
$$\mathbf{B}^{(m)} = -ikrot\mathbf{E} = E_0 \frac{w_0}{k} \left\{ \begin{array}{l} \ldots + \dfrac{\alpha}{k} \dfrac{\partial^2 U^{(m)}}{\partial x \partial y} + \dfrac{\beta}{k} \dfrac{\partial^2 U^{(m)}}{\partial y^2}, \\ \ldots - \dfrac{\alpha}{k} \dfrac{\partial^2 U^{(m)}}{\partial x^2} - \dfrac{\beta}{k} \dfrac{\partial^2 U^{(m)}}{\partial x \partial y}, \\ \ldots\ldots \end{array} \right\} e^{ikz}$$

a 2nd order derivative appears

2nd order derivative for B, ***vs.*** **1st order** derivative for E

B and **E** actuallly calculated **at the same order** with respect to **A**

LAGUERRE-GAUSS : a genuine "holey" beam ?



MAGNETIC ENERGY ON-AXIS !

V. Klimov *et al*, Opt. Express **2009**

calculation is for an **extreme focusing** ($w_0 \sim \lambda$ or possibly $kw_0 = 10$)

$$|B|^2 = E_0^2 \frac{16(p+1)(p+2)}{\pi(kw_0)^4} |\alpha \mp i\beta|^2 \neq 0$$

NANO-OPTICS

In optics, E and B are "identical" ***only if*** **sub-λ structure negligible** (*e.g.* ***standing wave!***)

Chirality often associated with **circular polarization**:

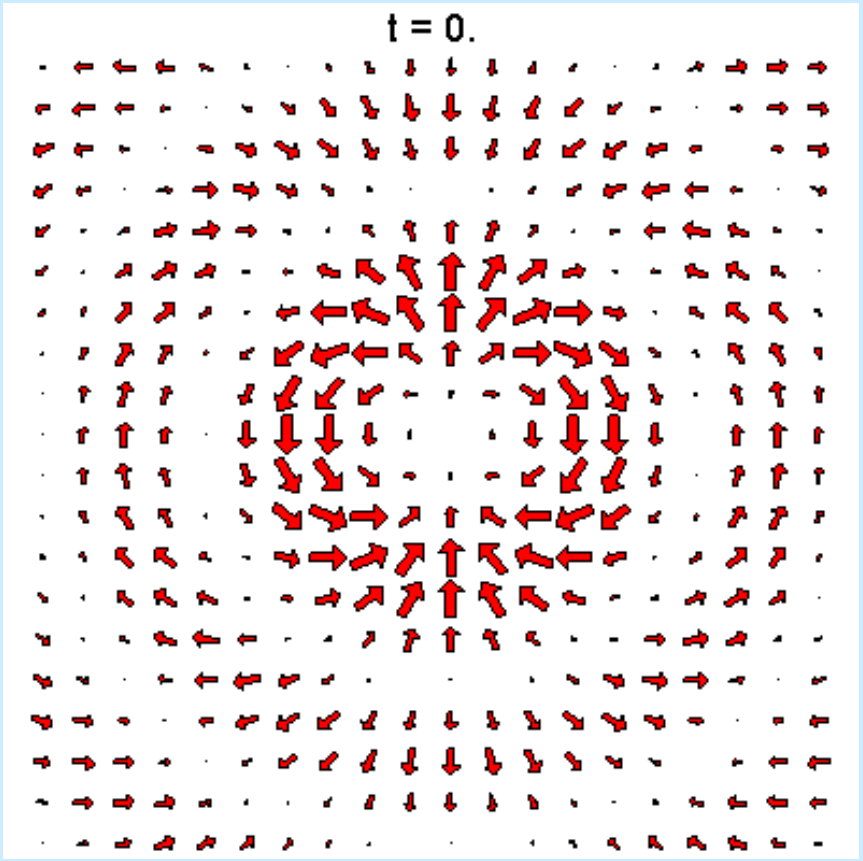
HERE, LOCAL structure of the field is of interest

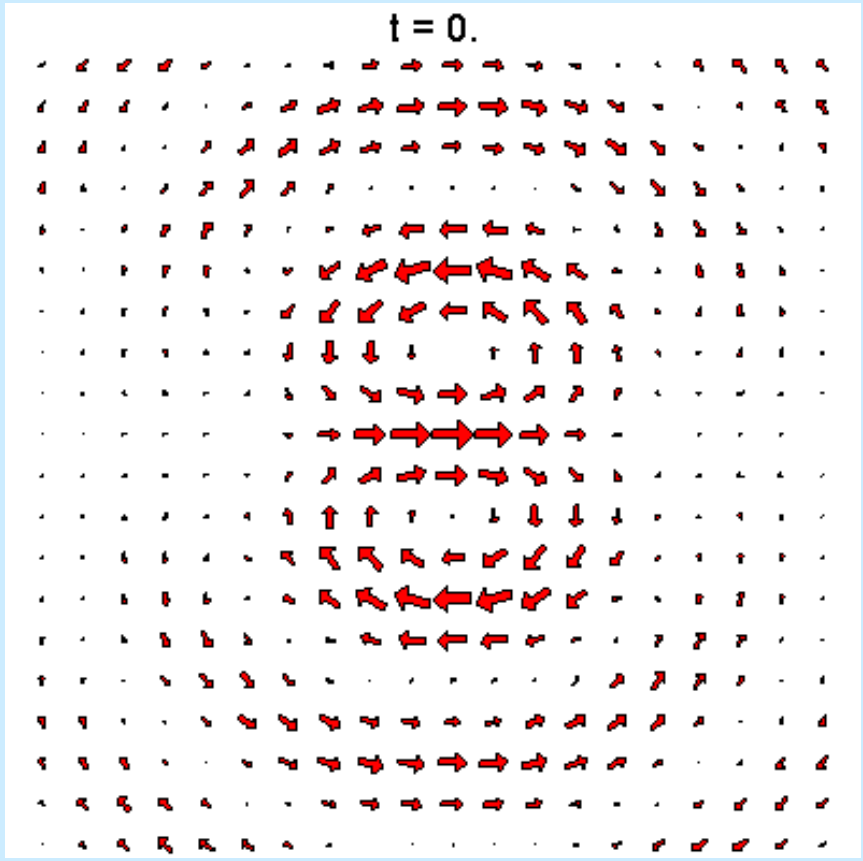
Distribution of **E**

$\sigma = -1$, $kw_0 = 6$

***l* = +2**

Distribution of **B**





locally, there is a circular polarization, according to ***l***

NEARLY FORBIDDEN transitions in **OPTICS**

To "feel" **B** w/o E : **M1** transition

to "feel" ∇**E** w/o E: **E2** transition

For a plane wave, there is a gradient **along λ**

(see also ***evanescent*** wave)

E2 or M1 typically weaker than E1 by a factor $(a_0/\lambda)^2 \sim 10^{-8}$

for E3 or M2 the factor is : $(a_0/\lambda)^4$…

Increasing a_0 **?** big molecules, or polymers ?

Atoms moving in a gas ?

Not good for a sub-λ control of positions

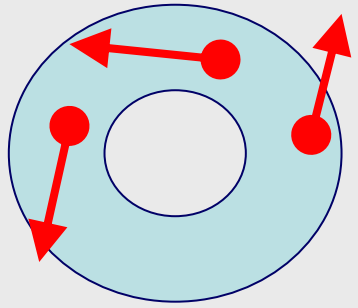
$\rightarrow$ transitions of atoms embedded in a matrix is better

Momentum Transfer from photon to atom

internal ***or*** external

*For an **ensemble** of particles*

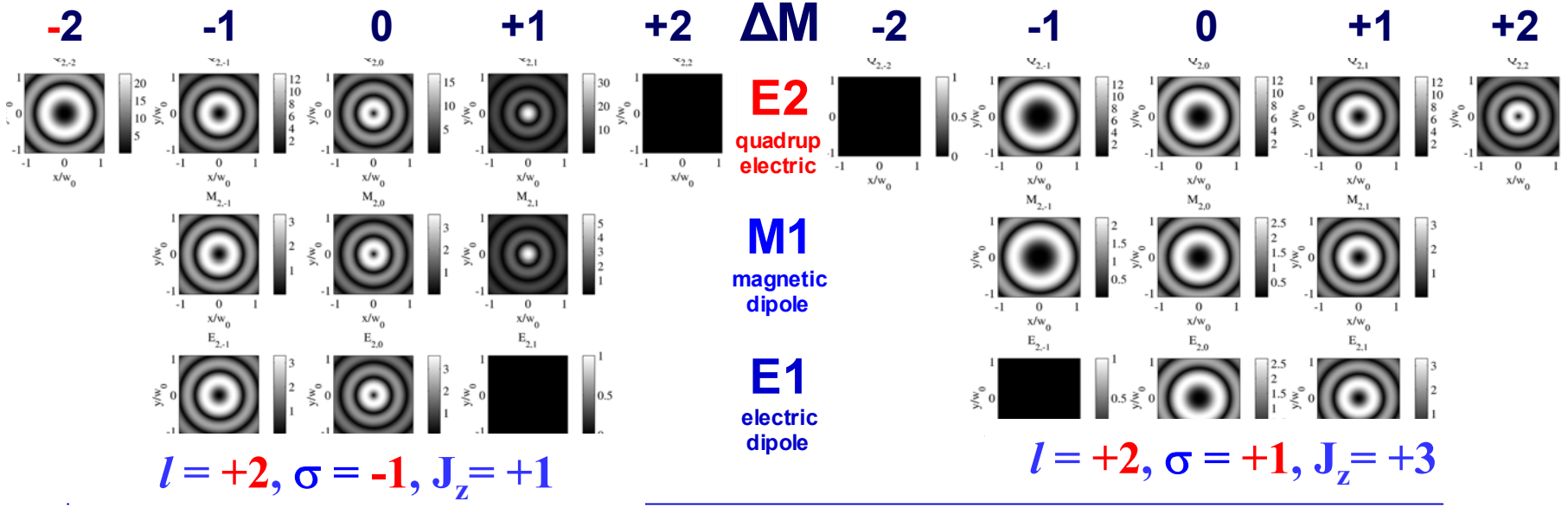
$\rightarrow$ rotation along the axial singularity



Mapping LG beams with E1, M1, E2 (Zeeman-sensitive)-detectors

focusing $kw_0=6$, p=6

S state (m=0) to P (m +0, ±1), or D (m = 0, ±1, ±2)

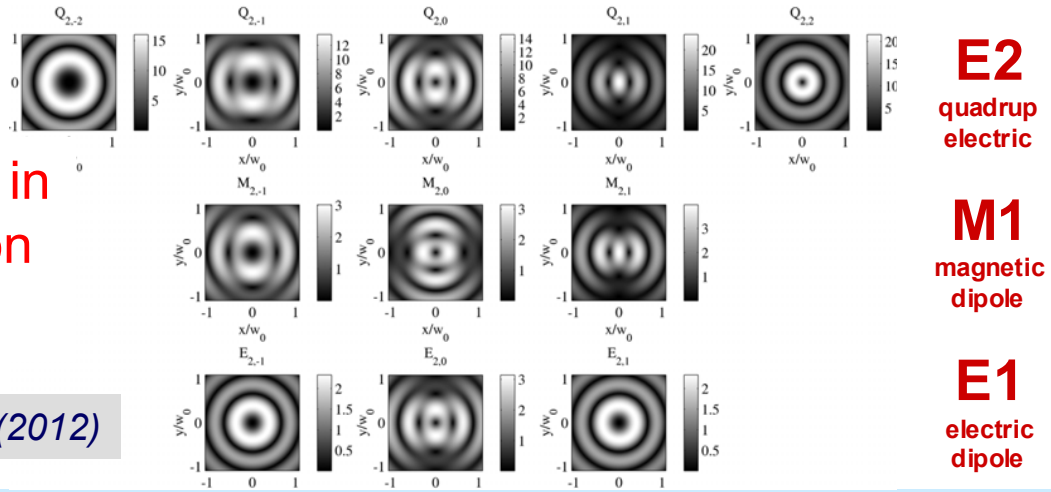


$l = +2, \sigma = -1, J_z = +1$

$l = +2, \sigma = +1, J_z = +3$

symmetry break in **linear** polarization

(σ=+1 **et** σ=-1)



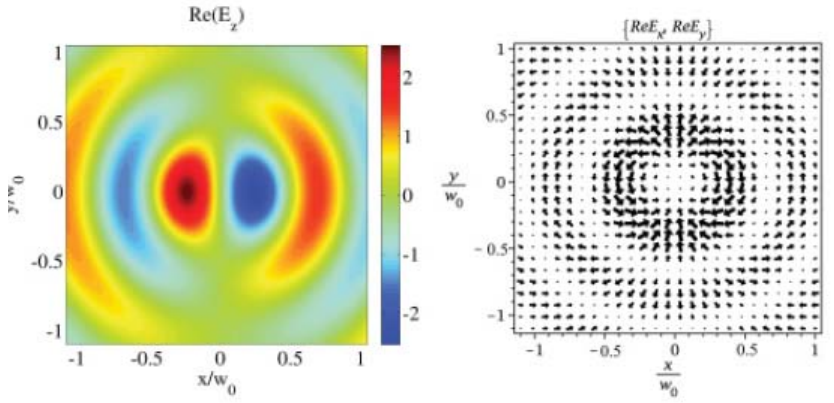
*V. Klimov et al. PRA **85** 053834 (2012)*

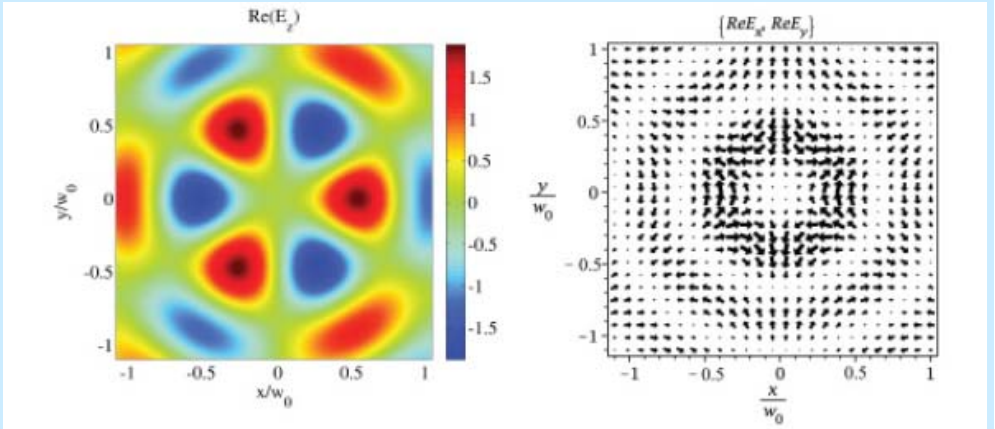
$l = +2, \sigma = -1$

~ *spin-orbit coupling*

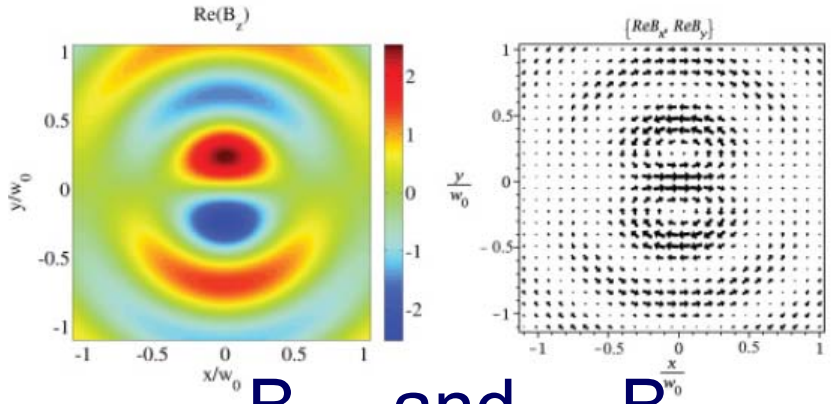
$l = +2, \sigma = +1$

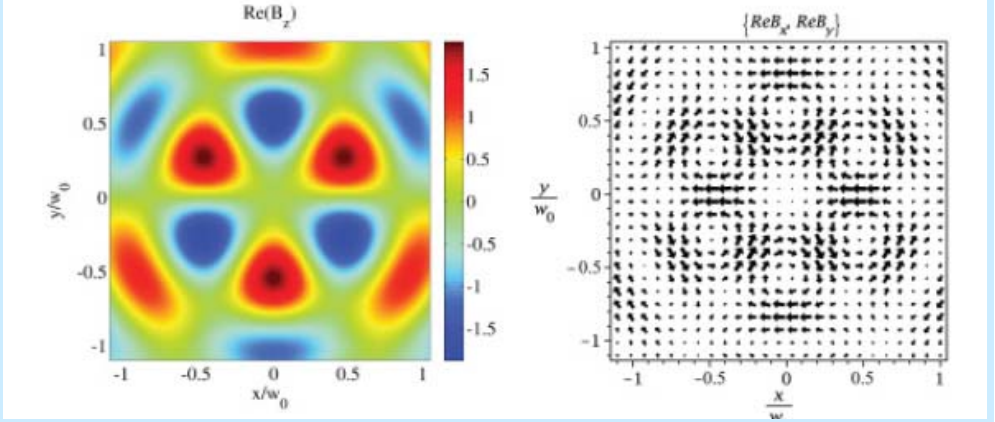
E_z and E_{trans}





B_z and B_{trans}





ternary symmetry, according to **total momentum** $l + \sigma$

LG chirality for chiral particle selectivity ?

PHYSICAL REVIEW A **71**, 055401 (2005)

Interactions of twisted light with chiral molecules: An experimental investigation

F. Araoka, T. Verbiest, K. Clays, and A. Persoons
KU Leuven, Laboratory of Chemical and Biological Dynamics, Celestijnenlaan 200 D, B-3001 Leuven, Belgium
(Received 21 January 2005; published 11 May 2005)

Abstract *"We experimentally demonstrate that helical* ***Laguerre-Gaussian light,*** *which carries orbital angular momentum* ***is not specific in its interaction with chiral matter.*** *Only circular polarization, associated with spin angular momentum, can engage with materials chirality."*

.....

"...this conclusion is only valid for linear optical processes and ***does not exclude chiral interactions for higher-order*** *optical processes."*

The answer is **NO**
when integrated on the whole beam

OAM and "Super-CHIRAL" sensitivity ?

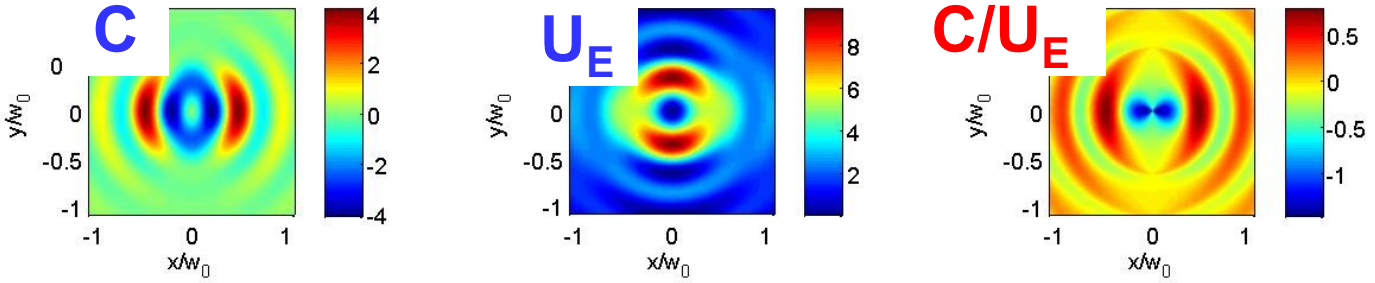
chiral molecule : the sensitive transitions are **E1** + ***M1***

C = $\varepsilon_0/2$ **E.**∇ **x E** + $1/2\mu_0$ **B.** ∇ **x B** (*i.e.* **E. B*),**

C/U_E : chiral sensitivity (*Tang and Cohen* PRL **104**, 163901 (2010))

The idea is to benefit of a local increase of B/E

l = 2
kw_0=6
p=6



LOCAL Chiral sensitivity is predicted only for a sub-λ scale

Calculations are to be extended: ***l, p, w_0***

Increasing *l* does not increase the gradient (λ ou $\lambda/2$)

Stongly Focused LG → NOT a *non-paraxial* "focused" LG
but some analogies !

August 15, 2007 / Vol. 32, No. 16 / OPTICS LETTERS 2357

Investigation of the center intensity of first- and second-order Laguerre–Gaussian beams with linear and circular polarization

Yoshinori Iketaki,[1,*] Takeshi Watanabe,[1] Nándor Bokor,[2] and Masaaki Fujii[3]

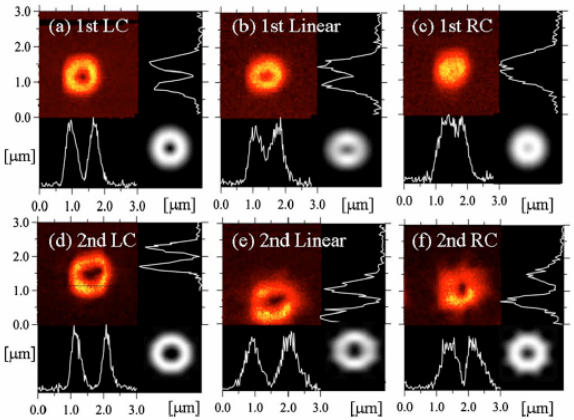


Fig. 4. (Color online) Measured intensity contour maps and profiles of the focused 1st- and 2nd-order LG beams: (a)–(c) 1st-order LG beam, (d)–(f) 2nd-order LG beam. The polarizations are (a) and (d) LC, (b) and (e) linear, (c) and (f) RC.

26 December 2005 / Vol. 13, No. 26 / OPTICS EXPRESS 10440

Investigation of polarization effects for high-numerical-aperture first-order Laguerre-Gaussian beams by 2D scanning with a single fluorescent microbead

Nándor Bokor

Department of Physics, Budapest University of Technology and Economics, Budapest 1111, Hungary
nandor.bokor@weizmann.ac.il

Yoshinori Iketaki, Takeshi Watanabe

Olympus Co., Ltd., 2-3 Kuboyama-cho, Hachioji-shi 192-8512, Japan

Masaaki Fujii

Chemical Resources Laboratory, Tokyo Institute of Technology, Yokohama 226-8503, Japan

Symmetry breaking occurs **also** for a linearly polarized paraxial beam **under strong** (lens) **focusing**
(*an effect of longitudinal polarization under focusing*)

A singular problem with LG beams :

Transfer of singularity

from one axis (*z*) to another axis (*z*') ?

This seems to work (NLO, FWM)

(exp^{ts} Tabosa group in Recife + L. Pruvost)

but approximately...

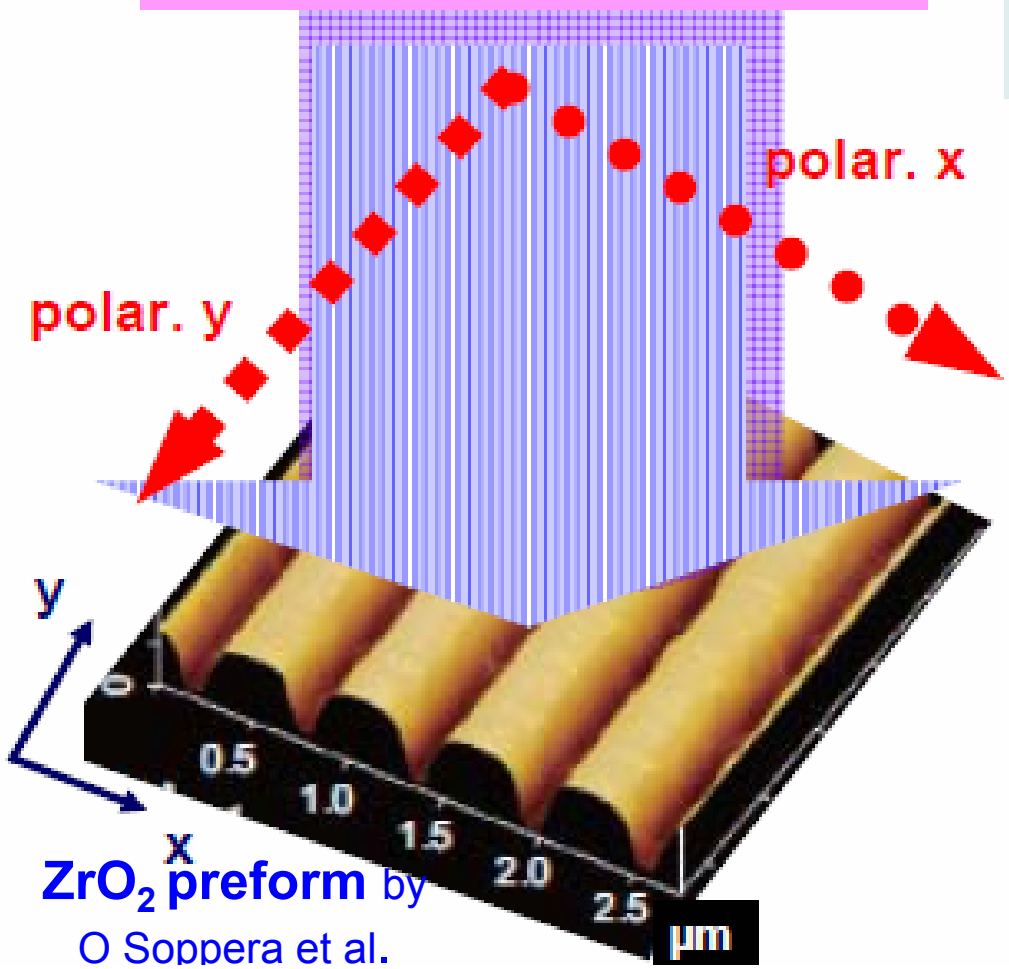
there are some impossibilities for 90 ° ..

and meaning if chirality...rotation by +/- 90°

Nano-etching or nano-smoothing by irradiating air

T Yatsui *et al.* **Light**: Science and Applications **5**, e16054 (2016)

irradiation He-Cd
325 nm, 8 mw/mm²
in air : $O_2 \rightarrow O^* + O$

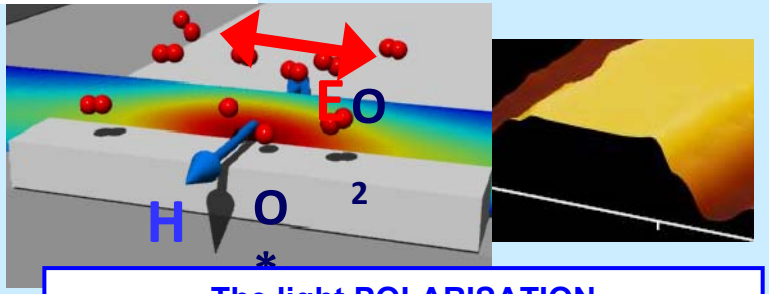


ZrO_2 preform by
O Soppera et al.

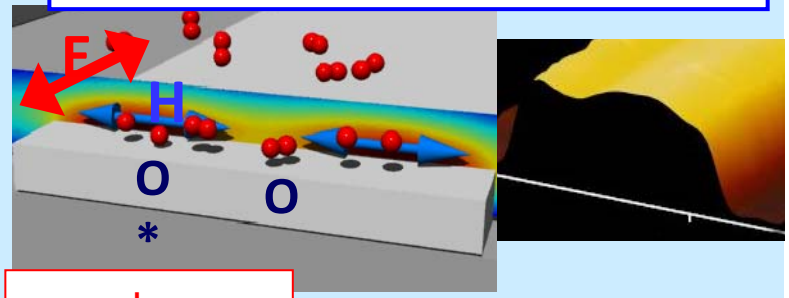
step : ~ 600 nm (450 + *150*)

depth : ~ 100 nm

polar X

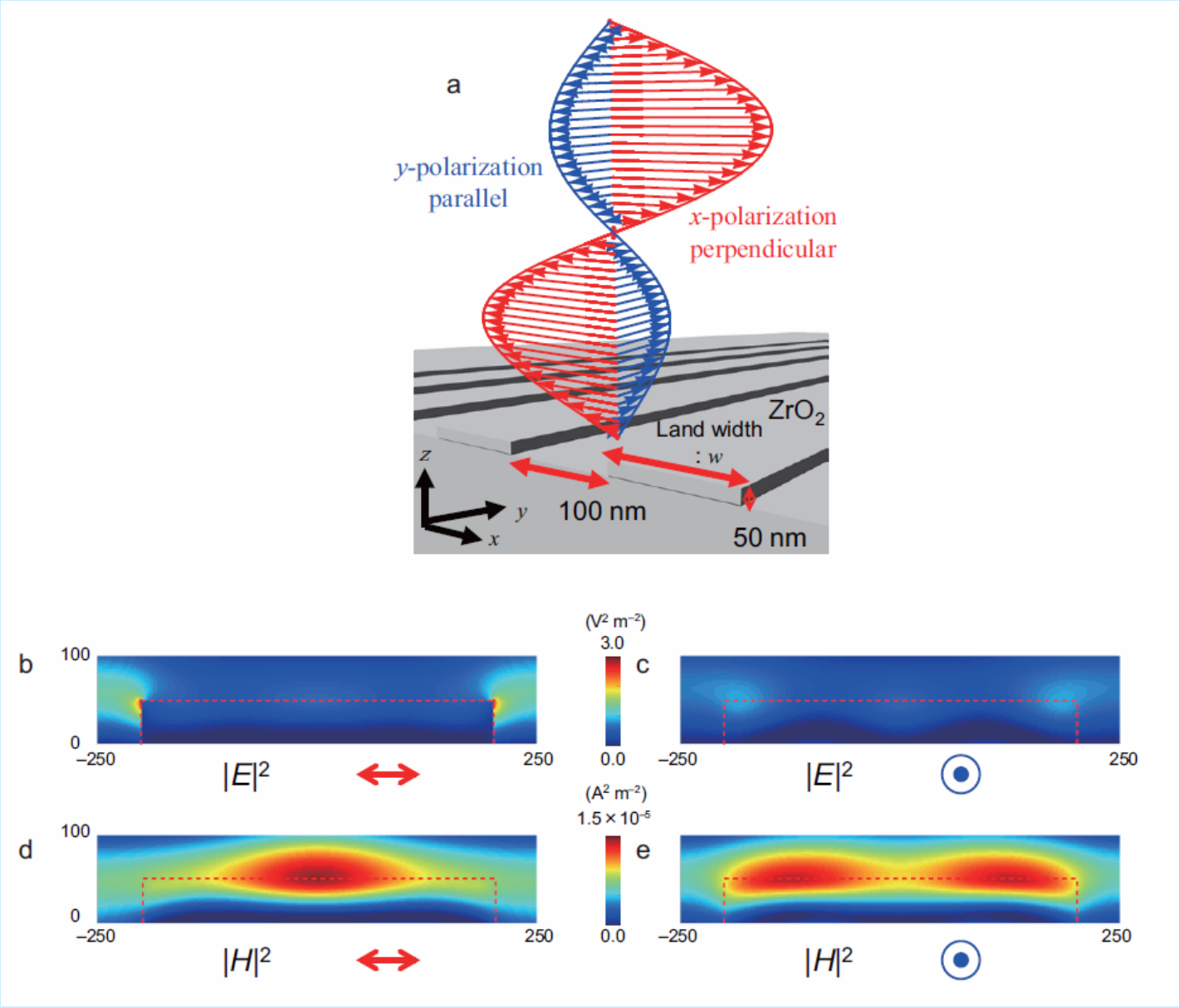


The light POLARISATION affects the induced STRUCTURE !



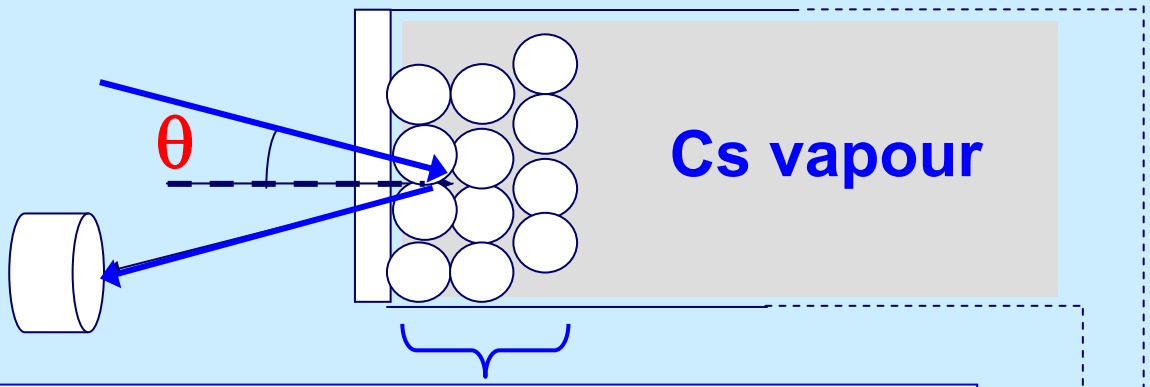
polar y

after 3 hours of irradiation



What happens if the initial structure has "some" chirality ?

Reflection spectroscopy at the opal surface



THIN LB opal - **interstices ~0.5µm**
vs. macroscopic cell

ARTIFICAL OPALS: arrangement of glass nanospheres
photonic crystal prepared by (self-organized) soft chemistry
(For us, a gas container, for gas confined in the interstices)

Reflection at interface → Reflection **spectroscopy**
sub-Doppler contribution (with FM)
Comparing **20** layers, **10** layers, **4, 3, 2** layers, and ..**1 layer**

HEXAGONAL DIFFRACTION ON A 1 LAYER OPAL

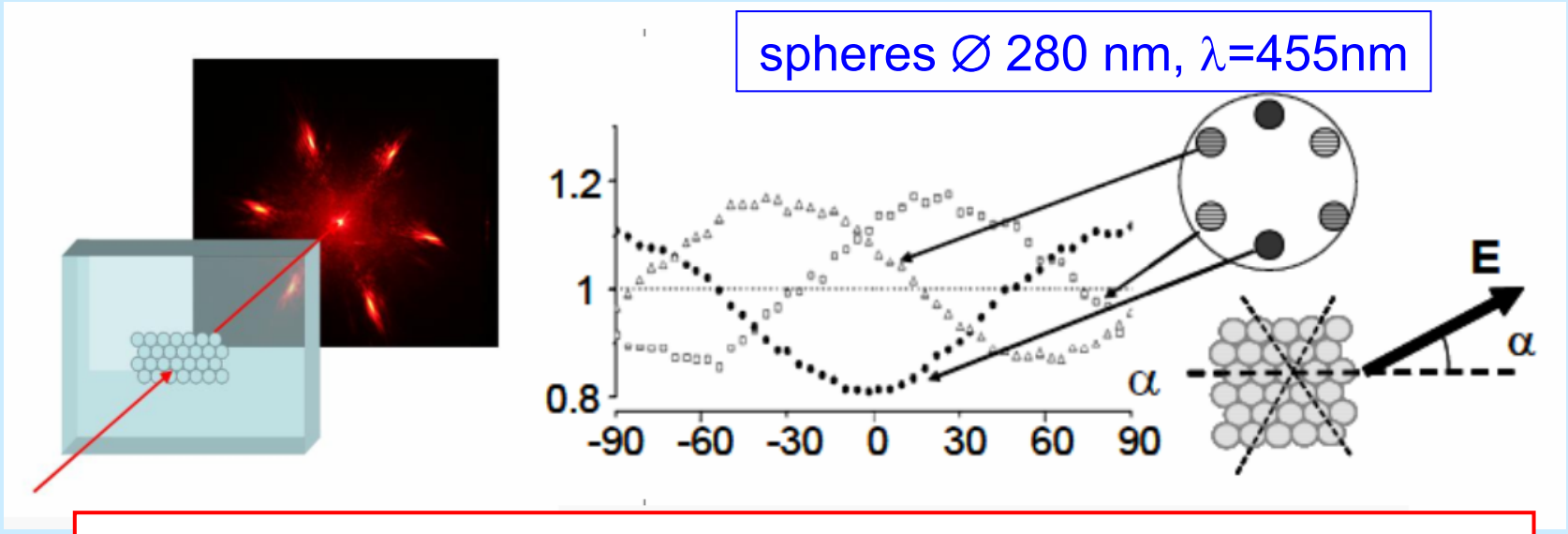
Artifical opals : (self-organized) arrangement of glass nanospheres

allows photonic crystal prepared by soft chemistry

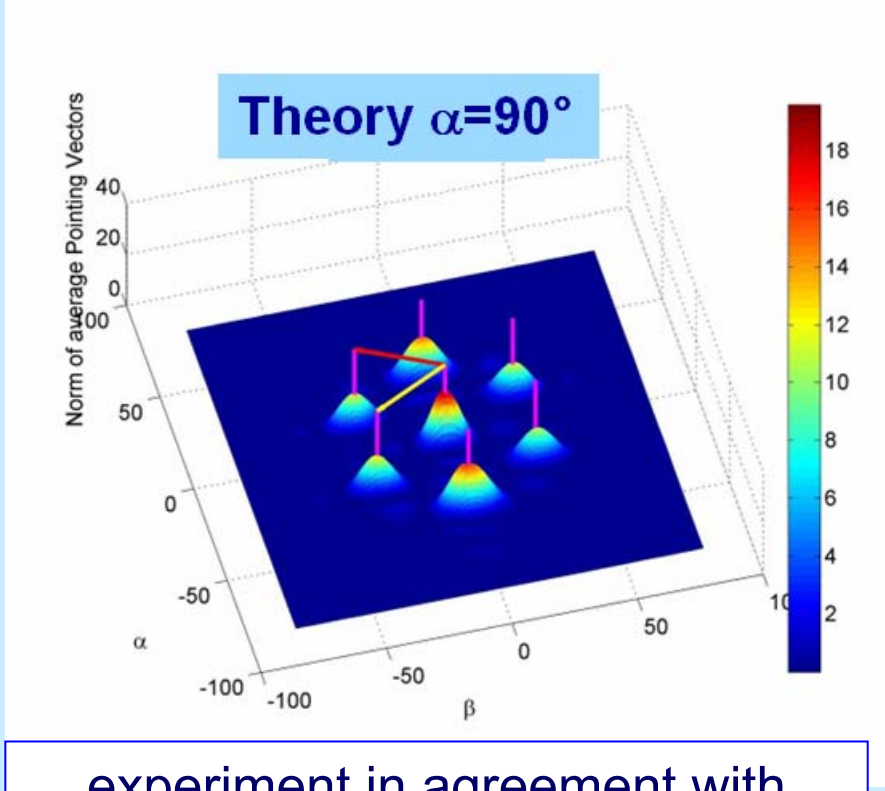
(For us, a gas container, for gas confined in the interstices)

"Bragg diffection", despite connected spheres ≠ nuclei of a crystal

Depolarisation effects usually neglected



The ternary symmetry **induces birefringence**



experiment in agreement with theoretical numercial calculation

I Zabkov, V Klimov, *et al, unpublished*

Future plans

replacing contacting spheres

by nanoetched gas containers:

hexagonal vs. square distribution by nanofabrication

Chiral distribution to be considered

and mixed with polar. anisotropy

Contributions by

J.R. Rios Leite (LG, + chirality) (UFPe Recife, Brazil)

V.V. Klimov (LG, + chirality) (Lebedev, Moscow)

M. Ducloy (LG)

T. Yatsui (M1 transitions) (Tokyo U.)

S. Tojo (M1 transitions) (Chuo U., Tokyo)

O.Soppera (ZrO_2 nanostructures) -IS 2M CNRS

E. Moufarej (opal)

I.Maurin (opal)

A. Laliotis (opal)

I. Zabkov (opal) (Lebedev, Moscow)

+ **informal discussions:** JWR Tabosa (Recife) , L Pruvost (Orsay), R Mathevet (Toulouse), O Emile and C Brousseau (Rennes)

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