Nonlinear optics determination of the symmetry group of a crystal using structured light

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We put forward a technique to unveil to which symmetry group a nonlinear crystal belongs, making use of nonlinear optics with structured light. We consider as example the process of spontaneous parametric down-conversion. The crystal, which is illuminated with a special type of Bessel beam, is characterized by a nonlinear susceptibility tensor whose structure is dictated by the symmetry group of the crystal. The observation of the spatial angular dependence of the lower-frequency generated light provides direct information about the symmetry group of the crystal.

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The spatial arrangements of atoms of substances that can present themselves in a crystalline structure is determined by the specific (point group or symmetry group) category [1] to which the crystal belongs. According to Neumann’s principle [2], or principle of symmetry, if a crystal is invariant with respect to a set of symmetry transformations, any physical property of the crystal is also invariant under such operations. Thus, the response of a crystal to optical, electric or magnetic stimulus can be used to predict general features of its structure, i.e., its symmetry group, allowing to unveil its presence in a determined spatial region of interest.

Crystallographic characterization is usually performed by diffraction techniques using X-rays [3], or material waves [4, 5] that interact with the atoms that are arranged in a certain pattern dictated by the symmetry group. By observing the spatial distribution of the photons or particles that come out from the crystal, one can obtain the sought-after information. In spite of the high importance of such techniques, sometimes they cannot be used, or its use is cumbersome and extremely difficult. Moreover, they are not immune to obstacles that may prevent a unique symmetry assignment to a given diffraction pattern. This can be due, e. g., to insufficient number of Bragg peaks owing to a finite instrument momentum range or spurious peaks arising from multiple scattering events. In these cases, a companion technique might help to identify the appropriate symmetry group of the crystal.

An alternative to diffraction techniques is the characterization of the crystal symmetry by its effects on the nonlinear optical response encoded in the susceptibility tensors [6–8]. These tensors are particularly useful because of their high sensitivity to both lattice and electronic symmetries. In general, the polarization induced in a material non-linear optical response encoded in the susceptibility tensors [6–8]. These tensors are particularly useful because of their high sensitivity to both lattice and electronic symmetries. In general, the polarization induced in a material when it is illuminated by an optical beam can be written as the Bloembergen expansion [9]

\[ P_i = \epsilon_0 \chi^{(1)}_{ij} E_i + \epsilon_0 \chi^{(2)}_{ijkl} E_i E_j + \epsilon_0 \chi^{(3)}_{ijklm} E_i E_j E_k E_l \ldots \]  

where \( E_i \) (i = x, y, z) are the components of the electric field, \( \chi^{(1)}_{ij} \) is the linear susceptibility tensor that determines the linear polarization response of the material, and \( \chi^{(2)}_{ijkl}, \chi^{(3)}_{ijkl} \ldots \) are nonlinear susceptibility tensors of different ranks responsible for the nonlinear polarization generated in the medium.

Here we show two main things. Firstly, that it is possible to perform complementary symmetry studies of nonlinear materials by using nonlinear optics processes besides second harmonic generation, such as spontaneous parametric down-conversion (SPDC). In SPDC, an intense pump beam with frequency \( \omega_p \) interacts with the atoms or molecules of a second order nonlinear crystal with nonlinear coefficient \( \chi^{(2)}_{ijk} \). In the process, a flow of paired photons is generated, the signal and idler, with central frequencies \( \omega_s \) and \( \omega_i \), such that \( \omega_s + \omega_i = \omega_p \).

Secondly, that instead of illuminating the nonlinear crystal with paraxial Gaussian beams that propagate along a myriad of different directions, as it is usually done in similar cases [2], it is more advantageous to choose as illuminating beam a non-paraxial optical beam. The use of a laser source with fast switching of its pointing direction might be technically cumbersome, and can severely limit the applicability of the method due to the need to control mechanical noise. However, the use of an appropriately designed non-paraxial beam allows to unveil the crystal symmetry group in a single shot experiment.

Under standard conditions [10], most SPDC configurations consist of a Gaussian pump beam that impinges normally onto the surface of the nonlinear crystal, which is endowed with a second order nonlinear susceptibility \( \chi^{(2)}_{ijk} \). The cut angle of the crystal is chosen to guarantee the fulfillment of the phase matching conditions, which are equivalent to the conservation of energy and momentum of the photons involved in the SPDC process. In most cases, the pump beam is linearly polarized. Under these circumstances, the distribution of wave vectors of the signal and idler photons, for given frequencies of the resulting photon pairs, is restricted to a single cone (in the case of type I SPDC) or to two cones (in the case of type II SPDC) with axes symmetrically arranged relative to the pump beam [11]. These conical distributions are observed for different crystal symmetries, so they do not bear useful information about the point group to which the crystal belongs. This is because in the configuration considered, the nonlinear process addresses only certain elements of the nonlinear susceptibility tensor.

We show that a different configuration can lead to a spatial distribution of the photon pairs that directly reflects the symmetry group of the crystal. For the sake of simplicity, we will consider the case of a uniaxial birefringent crystal with its optics axis parallel to the normal of its surface. The pump beam that illuminates the crystal is a vectorial Bessel mode [12, 13], prepared to guarantee a vectorial extraordinary character inside the nonlinear media. This can be done by choosing a transverse magnetic (TM) mode with its main propagation direction coincident with the axis of the nonlinear crystal.

A Bessel beam is formed by the superposition of plane waves with wave vectors confined in a cone and with a circular cylindrical symmetry on the angular spectrum. In this way, the cylindrical symmetry of the photon pairs produced in the SPDC process is directly broken by the intrinsic symmetry of the crystal, which manifests on the spatial distribution of the resulting photon pairs. The axicon angle of the Bessel vectorial mode is the parameter to be optimized for both the fulfillment of the phase matching conditions and to obtain a clear visibility of the characteristic crystallographic pattern built by the photon pairs.

Note that choosing a Bessel beam as a pump of the nonlinear process is equivalent to observing the crystal structure simultaneously for many different angles. In this configuration, SPDC (or any other nonlinear optical
process) is sensible to all the components of the nonlinear optical susceptibility tensor, including that along the main direction of propagation of the pump beam; that is due to the fact that TM modes acquire a significant component of its electric field along their main direction of propagation as they depart from the paraxial limit [13].

In this paper we consider vectorial monochromatic beams, i.e., beams with an electric field $\mathbf{E}(\mathbf{r}, t) = 1/2 \mathbf{E}_m(\mathbf{r}) \exp \left(-i\omega t\right) + c.c$ where $\omega$ is the angular frequency of the beam, $\mathbf{r}$ designates the spatial location, $t$ is time, $\mathbf{E}_m$ is the vectorial spatially-varying amplitude of the beam and $m$ is related to the orbital angular momentum of the beam.

In free space, the electric field of a transverse-magnetic (TM) Bessel beam of order $m$, which is an exact solution of Maxwell’s equations, can be written [12] as a sum of plane-waves with equal amplitude and wavevector confined in a cone around the $\hat{z}$ axis, the main direction of propagation of the beam, with the so called axicon angle $\varphi_a$, i.e.,

$$\mathbf{E}_m(\mathbf{r}) = E_0 \int d\mathbf{p} \mathbf{e}_k \exp \left(ik_z z + i\mathbf{p} \cdot \mathbf{r}_\perp + im\varphi_p\right)$$

(2)

where $\mathbf{e}_k = \hat{k}(\mathbf{k} \cdot \hat{z}) - \hat{z}$ is the polarization of each wave with wavevector $\mathbf{k}$, the longitudinal wavenumber is $k_z = (\omega/c) \cos \varphi_a$, and the transverse wavevector writes $\mathbf{p} = (\omega/c) \sin \varphi_p (\cos \varphi_p \hat{x} + \sin \varphi_p \hat{y})$. $\varphi_p$ is the angle between the transverse wavevector $\mathbf{p}$ and $\hat{z}$, and can take any value between 0 and $2\pi$. Notice that even though the polarization of each $\mathbf{k}$-wave is perpendicular to $\mathbf{k}$ [13], the superposition of all $\mathbf{k}$-waves yields a beam with a non-zero field component along the main direction of propagation ($\hat{z}$). For paraxial beams ($\varphi_a$ small) and $m = 0$, one obtains the so-called radial modes [12, 15].

In experiments, one never generates such an ideal beam. However, one can generate vectorial Bessel beams close to the one described by Eq. (2) by generating superpositions of scalar Bessel modes with the proper topological charge and polarization [10]. Under these experimental conditions, the wavevectors $\mathbf{k}$ are no longer confined to a cone of angle $\varphi_a$, with transverse wavevector $\mathbf{q}$, but instead the wavevectors spread narrowly around a central value, $\mathbf{q} + \Delta \mathbf{q}$, or equivalently, $\varphi_a + \Delta \varphi_a$, with $|\Delta \mathbf{q}| = |\mathbf{k}|\Delta \varphi_a$.

The Hamiltonian of interaction of SPDC is [17]

$$\hat{\mathcal{H}}(t) = \epsilon_0 \int_V dV \int d\mathbf{k}_p \int d\mathbf{k}_s \int d\mathbf{k}_i \chi^{(2)} \mathbf{E}_p(t, \mathbf{r}; \mathbf{k}_p) \mathbf{E}^*_\perp \mathcal{E}_\perp \mathbf{E}^* \mathcal{E}_\perp(t, \mathbf{r}; \mathbf{k}_s) \mathbf{E}_i \mathcal{E}_\perp(t, \mathbf{r}; \mathbf{k}_i) + h.c.$$  

(3)

where $V$ is the volume of interaction and $\chi^{(2)}$ is the nonlinear tensor that characterizes the nonlinear response of the material.

Let us consider as example a type I (eeo) SPDC process in an uniaxial crystal [18]. A configuration that will give us information about the symmetry of the crystal is the one where the pump beam propagates inside the nonlinear crystal along the optical axis ($\hat{c} = \hat{z}$). The signal and idler waves propagate as ordinary waves. For the case of an intense classical extraordinary pump beam that generates ordinary signal and idler photons, the pump beam, and the electric field operators for signal and idler photons write [10]

$$\mathbf{E}_p^\perp(t, \mathbf{r}; \mathbf{q}_p) = E_0 \mathbf{e}_p(\mathbf{q}_p) e^{im\varphi_{op}} e^{ik_p^s(\mathbf{q}_p) z + iq_p \mathbf{r}_\perp},$$

$$\mathbf{E}^\perp_\perp(t, \mathbf{r}; \mathbf{p}) = iN_s \mathbf{e}_s(\mathbf{p}) a_s^\dagger(k_s^s(\mathbf{p}), \mathbf{p}) e^{ik_s^s(\mathbf{p}) z + ip \mathbf{r}_\perp},$$

$$\mathbf{E}_i^\perp(t, \mathbf{r}; \mathbf{q}) = iN_i \mathbf{e}_i(\mathbf{q}) a_i^\dagger(k_i^s(\mathbf{q}), \mathbf{q}) e^{ik_i^s(\mathbf{q}) z + iq \mathbf{r}_\perp},$$

(4)

where $N_s$ and $N_i$ are normalization factors,

$$k_p^s(\mathbf{q}_p) = \sqrt{\left(\frac{\omega_p}{c}\right)^2 \epsilon_{op} - \left(\frac{\epsilon_p}{\epsilon_{op}}\right)|\mathbf{q}_p|^2},$$

$$k_s^s(\mathbf{p}) = \sqrt{\left(\frac{\omega_s}{c}\right)^2 \epsilon_{os} - |\mathbf{p}|^2},$$

$$k_i^s(\mathbf{q}) = \sqrt{\left(\frac{\omega_i}{c}\right)^2 \epsilon_{oi} - |\mathbf{q}|^2},$$

$\epsilon_{op, os, oi}$ are the ordinary relative permittivities inside the crystal of the pump, signal and idler waves, $\epsilon_{ep, es, ei}$ are the corresponding extraordinary ones, and

$$\mathbf{e}_p(\mathbf{q}_p) = \left[\frac{e_p^2}{\omega_p^2 \epsilon_{op}} \mathbf{k}_p \cdot \hat{z} - \hat{z}\right],$$

$$\mathbf{e}_s(\mathbf{p}) = \mathbf{k}_s \times \hat{c}, \quad \mathbf{e}_i(\mathbf{q}) = \mathbf{k}_i \times \hat{c}$$

(5)
calculating the first-order solution of the Schrödinger equation, i.e.,

$$|\Psi(t)\rangle = |\text{vac}\rangle - \left(\frac{i}{\hbar}\right) \int_{-\infty}^{t} dt' \mathcal{H}(t') |\text{vac}\rangle_s |\text{vac}\rangle_i$$

(6)

The interaction Hamiltonian is effectively zero when the classical beams that pump the nonlinear process are zero, so that, the time of integration can be extended to $t = \infty$.

The quantum state of the down-converted photons at the output face of the nonlinear crystal, neglecting for the sake of simplicity the contribution from the vacuum term can be written as

$$|\Psi\rangle \sim \int d\mathbf{p} d\mathbf{q} F(\mathbf{p}, \mathbf{q}) a_\mathbf{p}^\dagger(k^z_s, \mathbf{p}) a_\mathbf{q}^\dagger(k^z_z, \mathbf{q}) |\text{vac}\rangle$$

(7)

where

$$F(\mathbf{p}, \mathbf{q}) = \sum_{l,m,n} \chi^{(2)}_{lmn} [\mathbf{e}_p(\mathbf{p} + \mathbf{q})]_l [\mathbf{e}_s(\mathbf{p})]_m [\mathbf{e}_i(\mathbf{q})]_n$$

$$\times \text{sinc} \left(\frac{\Delta k_z(\mathbf{p}, \mathbf{q}) L}{2}\right)$$

$$\times \exp \left[\frac{i k^p(\mathbf{p} + \mathbf{q}) + k^s(\mathbf{p}) + k^i(\mathbf{q})}{2} L\right]$$

(8)

and $\Delta k_z(\mathbf{p}, \mathbf{q}) = k^z_s(\mathbf{p} + \mathbf{q}) - k^z_s(\mathbf{p}) - k^z_z(\mathbf{q})$.

It is important to remind here the crucial role that the tensorial character of $\chi^{(2)}$ plays in the SPDC configuration considered. Most experiments that make use of SPDC, due to the paraxial character of the pump, can be described using an effective nonlinear index [13] that writes $\chi^{(2)}_{eff} = (\epsilon_{p})_\perp [\chi^{(2)}_{e}]_\perp (\epsilon_{s})_\perp$, where $\epsilon_{p,s,i}$ are the linear polarizations of the pump, idler and signal photons, respectively. The flux of down-converted photons depends on the magnitude of the effective index. The vectorial structure of the electromagnetic field defines its polarization and, consequently, affects its total angular momentum. Thus, non paraxial pump beams are expected to yield interesting results determined by the structure of $\chi^{(2)}_{lmn}$ and its relation to the angular momentum content of the down-converted photons [19].

In general, $F(\mathbf{p}, \mathbf{q})$ describes an entangled state in the polarization and transverse wavevector degrees of freedom. This entanglement takes place both when considering the signal and idler photons as the two subsystems of the whole system, and when considering the two degrees of freedom, polarizations and transverse wavevectors, as the corresponding subsystems.

The flux rate $R_{si}(\mathbf{p}, \mathbf{q})$ of detections of a signal photon with transverse wavevector $\mathbf{p}$ in coincidence with an idler photon with transverse wavevector $\mathbf{q}$ (coincidence detection), or equivalently, a signal photon that propagates inside the crystal along the direction $(\theta_s, \varphi_s)$ with $\theta_s = \tan^{-1} [|\mathbf{p}|/\sqrt{(\omega_s/c)^2 t_0 - |\mathbf{p}|^2}$ and $\varphi_s = \cos^{-1} q_x/|\mathbf{p}|$ and an idler photon that propagates along the direction $(\theta_i, \varphi_i)$ with $\theta_i = \tan^{-1} [|\mathbf{q}|/\sqrt{(\omega_i/c)^2 t_0 - |\mathbf{q}|^2}$ and $\varphi_i = \cos^{-1} q_x/|\mathbf{q}|$ is

$$R(\mathbf{p}, \mathbf{q}) = |F(\mathbf{p}, \mathbf{q})|^2$$

(9)

while the flux rate $R_s(\mathbf{p})$ of signal photons detected with transverse wavevector $\mathbf{p}$ (singles detection) writes

$$R_s(\mathbf{p}) = \int d\mathbf{q} R(\mathbf{p}, \mathbf{q}) = \int d\mathbf{q} F(\mathbf{p}, \mathbf{q})^2.$$  

(10)

We illustrate these results, and how they help to determine the symmetry group of the crystal, by showing some examples. In Fig. 1, we exemplify the angular dependence of the flux rate of signal photons (singles detections as given by Eq. (10)) for a variety of nonlinear crystals with different crystallographic symmetries. The numerical simulations consider the adequate Sellmeier equations and the crystal birefringent properties. For small values of the transverse wave vector of the pump beam ($|\mathbf{q}_p| \leq 0.01 \mu m^{-1}$ for most of the crystals considered here, which corresponds to an angle $\varphi_a \ll 0.0097$), the angular spectrum is always formed by a set of concentric rings similar to those observed for $|\mathbf{q}_s| \leq 0.5 \mu m^{-1}$ in Fig. 1(a). As $|\mathbf{q}_p|$ increases the down-converted photons are emitted in a wider region in transverse wavevector space, and the symmetry of the crystal becomes more evident. Properties such as the radius of the concentric rings, the values of $|\mathbf{q}_p|$ at which the visibility of the symmetry of the crystal is significant, and the maximum value of $R_s$, could be used to determine some characteristics of the linear susceptibility tensor.
and the second-order nonlinear susceptibility tensors. For instance, KDP and CsH$_2$AsO$_3$ belong to the same point group, 42 m, but they have slightly different values of $\epsilon_o$, $\epsilon_e$ and the relevant components of $\chi^{(2)}$. This manifests in general common properties between the angular spectrum of both crystals, but there are still measurable differences for small values of $q_s$ due to a higher sensitivity of CsH$_2$AsO$_3$ to the electric field component along the $z$-axis.

The coincidence detections, given by Eq. (9) also provide useful information about the symmetry group of the medium under investigation. A direct calculation shows that for $|q_p| \ll \omega_p/c$, the detection of a signal photon with transverse wavenumber $p$ is accompanied by the detection of an idler photon that is confined to a ring with radius $|q| \sim |q_p|\epsilon_o/\epsilon_e$ around $q = -p$. As $q_p$ increases, the symmetry of the crystal becomes more visible in the angular distribution of idler photons, the flux rate becomes inhomogeneous along the ring with a structure that depends on the symmetry of the crystal, and which is compatible with the conditional generation of stationary Bessel photons. That is, photons with an angular spectra resulting from superpositions of $m$ and $-m$ Bessel modes. In Fig. 2, this effect is illustrated for various types of crystals.

Summarizing, we have shown that, under the adequate experimental set up, the correlations of the photons emitted in an SPDC process, contain crystallographic information that can be accessed by measurements of the SPDC angular spectra and the conditional angular spectra. It is expected that a complete characterization of the twin photons correlations as a function of the properties of the structured pump beam could be used to obtain precise measurements of the first and second order electric susceptibility tensors. Notice that the benefits arising from structured pump beams for nonlinear crystallography should not be restricted to the SPDC process. In fact, nonlinear harmonic generation crystallography [5] with Bessel beams could be an alternative to rotational anisotropy measurements [7], since using Bessel modes as a pump is equivalent to simultaneous measurements along different directions.

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FIG. 1. Angular dependence of the flux rate of signal photons detected [singles detection given by Eq. (10)] for type I degenerate SPDC ($\lambda_s = \lambda_i = 2\lambda_p$). The wavelength of the incident TM Bessel mode is $\lambda_p = 407$ nm. The angle of the cone that characterizes the illuminating pump Bessel beam is $\sin \varphi_a = 0.0097$. We also consider a spread of directions $\Delta \varphi_a \sim 0.0004$ rad. The crystal length is taken as $L = 1$ mm. (a) CdSe: point group 6mm; (b) KDP: point group 42m; (c) GaSe: point group 62m; (d) HgGa$_2$S$_4$: point group 4; (e) HgS: point group 32; (f) LiNbO$_3$: point group 3m
FIG. 2. Flux rate of idler photons detections in coincidence with the signal photons with the transverse wavevector $\mathbf{p}$ that maximizes $R_s$. We consider type I degenerate SPDC in a nonlinear crystal of length $L = 1\text{mm}$: (a) GaSe; (b)BBO; (c)LiNbO$_3$ and (d) KDP. The incident beam is a Bessel TM mode with wavelength $\lambda = 407\text{nm}$, $|\mathbf{q}_p| = 0.15\mu\text{m}^{-1}$ for (a-c) and $|\mathbf{q}_p| = 0.2\mu\text{m}^{-1}$ for (d). Again, we consider an spread of values of $\mathbf{q}_p$ of $\Delta \mathbf{q}_p = 1/150\mu\text{m}$. 

(a) \hspace{3cm} (b) 

(c) \hspace{3cm} (d)