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Bulk cylinders and spheres: from shaping to use for linear and nonlinear optics

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Abstract: Bulk crystals cut and polished as spheres or cylinders are unique tools for measuring the angular distribution of any linear or nonlinear optical property as well as for designing widely and continuously tunable parametric devices. The aim of this article is to describe the specific techniques of shaping such high quality samples, and their use in methods of optical measurements and devices that we implemented.

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1. Introduction

The classical methods of measurement of linear and nonlinear optical properties of crystals require cutting many samples of several centimetres size, which is not always compatible with the early stages of new materials growth. Furthermore, these methods use

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parallelepipeds or prisms, then the full angular distribution of optical properties cannot be determined. However it is of prime importance for many optical properties as the absorption coefficient, the spontaneous or stimulated emission cross sections, the nonlinear frequency conversion phase-matching directions, or the effective nonlinear coefficients, as examples. It is why we developed a new corpus of methods using only one sample of few millimetres size shaped as a sphere or a cylinder. For that purpose, we conceived specific techniques of cutting and polishing crystals as spheres or cylinders, the considerations of crystallographic orientation being of prime importance in this framework. Such geometries are of high degree of symmetry, enabling under rotation the propagation of a laser beam in any direction of the crystal. Thus the angular distribution of any optical property can be directly measured with the highest accuracy, improving the studied crystal engineering for its use in lasers and parametric devices.

The goal of this article is first to describe in details our original techniques of shaping, which is done for the first time. Then we show the different kinds of measurements, and associated accuracies, that can be performed in spheres using methods of optical measurements that we implemented. Finally, we demonstrate the interest of cylindrical crystals for designing widely and continuously tunable optical parametric devices.

2. Qualification and selection of the cutting volume

Most techniques probing the volume quality are in transmission geometry; hence the first task is to realize a slab with parallel faces for inspection. Parallel faces can be made either by lapping or cutting, the later being preferred for large sizes samples. The desired orientation in the sample is first aligned with a laser beam set in autocollimation, in order to create the first face. If this orientation has been previously defined by X-ray diffraction, then the orientation by autocollimation is set by using a goniometer and a reflecting reference attached to the sample holder. Then this first face is used as a reference, to create the second one. In such a procedure, the two faces are parallel within 0.05°, and the accuracy of their crystallographic orientation is within 0.01°. They are polished down to a surface roughness root mean squared (r.m.s) of 10-nm-r.m.s. Among the various techniques available to control the quality of the selected volume, one is polarized light microscopy to evidence major defects, as polycrystals, twinning, or fractures. Then more sensitive techniques, such as shadowgraphy or schlieren [1] may be used to reveal fainter refractive index variations due to sector boundaries typically, strains, or chemical inhomogeneities in the bulk crystal.

3. Cylinder shaping

The process of shaping and polishing crystals into cylinders is schematized in Fig. 1.

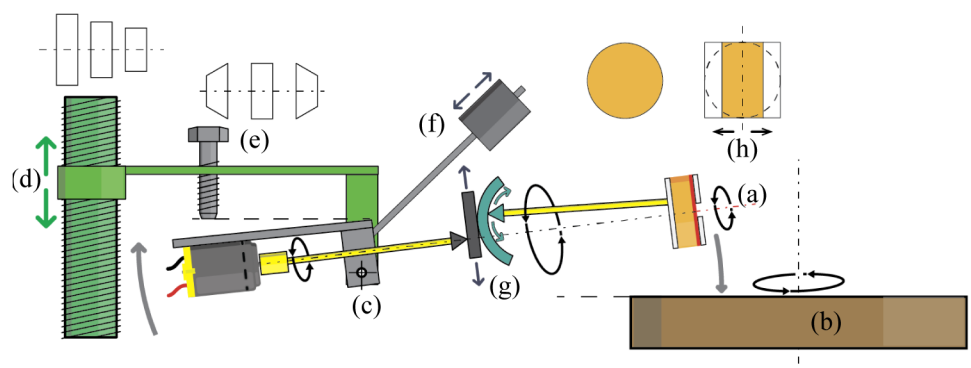

Fig. 1. Apparatus designed to shape crystals into cylinders with a polished rim.

It consists in bringing a crystal rotating around a defined axis (a), in contact with a series of lapping and polishing plates (b). This rotation axis defines the revolution axis of the cylinder. Therefore the first requirement is to realize a reference face in the crystal

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perpendicular to this axis: in most cases, it corresponds to a principal crystallographic direction, so that the reference face is the associated crystallographic plane. In such situations, X-ray diffraction enables the orientation of the cut reference face with an accuracy of +/– 0.01°. The cylindrical shape is achieved through a progressive lapping of the sample rim using our homemade shaping apparatus shown in Fig. 1. At first, each prominent part is eroded keeping the sample static. When an obtuse polygonal geometry is achieved, the sample is rotated at about 10 rpm around the revolution axis (a). The whole sample/rotation device is mounted on a hinge (c) to accommodate for the initial variations in sample lateral dimensions. The amount of excess material to be removed is fixed by the joint effect of the translation (d) and the adjustment screw (e). The later serves as the hinge end stop. The pressure of the sample on the plate (b) is fixed through adjustable counterweights (f). When the shape has converged to a cylinder, the final diameter is finely tuned by fixing the distance between the revolution axis (a) and the lapping plate (b) thanks to the fine threaded vertical translation (d). The precise orientation of the cylinder reference face with respect to the revolution axis (a) is one of the key point of the process. Two cradles (g) allow the cylinder reference face to be set perpendicular to the revolution axis (a). Furthermore, two crossed translations (g) insure that this axis goes through the sample center in order to maximize the size of the final cylinder. Another key point is the orientation of the reference face that has to be set perpendicular to the plate (b). For that, the (d) translation is set high and the reference face is brought in autocollimation with a laser beam parallel to the lapping plate thanks to the adjustment screw (e). The tuning of screw (e) is critical as it controls the final geometry of the objet: cylindrical or with a conical tapper. The mounting of the sample to be shaped is the last important point. The sample is sandwiched between two 1-mm-thick glass slabs stuck on its parallel faces. This mounting stiffens very thin samples (<1mm), strengthens those with poor mechanical properties, lessens the effects of hardness anisotropy, which may lead to elliptical shapes, and limits edge beveling on the final cylinder.

This procedure enables producing cylinders with a revolution axis normal to the reference face within 0.1°, an acylindricity relative to the diameter below 0.1%, and a rim polished to a roughness of about 10 nm r.m.s. A partial, *i.e.* truncated, cylinder can be also interesting when a limited angular range is sufficient regarding the considered optical property, so that a longer diameter can be obtained. It can be realized from a rectangular sample with its long axis along the median direction of interest. For that, in the process described above, glass parts making up for the full cylinder are glued on each side of the crystal of same thickness. In this case the lateral centering of the crystal (h) on the rotation axis (a) is critical to avoid making an asymmetrical partial cylinder. Several examples of cylinders cut using this process are shown in Fig. 2.

Fig. 2. Examples of polished full or partial cylinders ready for optical studies.

4. Sphere shaping

The method introduced below allows to shape optically polished spheres of 3 to 10 mm in diameter. Like cylinders, they are shaped by the progressive abrading of the rotating sample, from five main steps illustrated by Figs. 3. No specific orientation of the starting sample is requested, which will be determined *a posteriori* by X-ray diffraction.

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(a) (b) (c) (d) (e)

Fig. 3. Main steps (a), (b), (c), (d) and (e) for shaping and polishing a sphere from a cube.

The first step of the process consists in shaping a sample as a cube (Fig. 3(a)) with 6 faces perpendicular to each other and of same size. This last request will facilitate the centering of the object on the successive rotation axes and control the isotropic removal of material toward the spherical shape. There is not need to cut the cube of Fig. 3(a) oriented. It is stuck on a pin on one of its faces, after bevelling all the edges with a template (Fig. 3(b)). Then the cube is mounted on a motorized rotation axis. It is centered on that axis thanks to two crossed translations. The sample is slowly rotated with a speed between 10 and 20 rotations *per* minute (rpm) to remove the linear edges as shown Fig. 3(c)-3(d). Concave hemispherical templates covered with abrasive suspension are manually put in contact with the rotating sample. The size of the hemisphere template is gradually reduced, so that the shape of the sample is changed from the cube diagonal down to the final sphere diameter, the corresponding abrasive particle size being reduced progressively. To converge toward a spherical shape, the sample has to be rotated around many different directions, so that the pin has to be unstuck and restuck many times. The remaining facets also serve to control that the shape is converging to a sphere. Actually, if the removal of matter is isotropic, which can be checked under stereomicroscope, the facets should all be of the same size with perfectly circular perimeters (Fig. 3(c)-3(d)). Once all facets have vanished, the spherical shape is achieved with a typical asphericity below 1% (Fig. 3(e)). It is then polished with stretched cloths and abrasive of small particle sizes to reach few nanometers surface roughness (rms).

The polished sphere sample is then stuck on a goniometer head in order to be oriented as requested by optical measurements. The orientation of the sphere is generally determined by the polychromatic Xray Laue technique. It may be necessary to make orientation adjustments greatly exceeding the goniometer head capacity, in order to stuck the sphere in the desired crystallographic orientation. In that case, the sphere has to be transferred from the initial goniometer head to that used for the optical measurements. The two goniometer heads are then mounted together on a horizontal spindle, the initial one being fixed and the receiving one mounted on a horizontal rotation circle. Using this rotation and taking into account the orientation of the crystallographic axes, the sphere is transferred and stuck oriented on the receiving goniometer head. The orientation of the crystallographic axes of the sphere are known with an accuracy better than 0.1°. Several shaped spheres mounted on a goniometer head are shown in Fig. 4.

Fig. 4. Examples of polished spheres mounted on goniometer heads for optical measurements.

5. Optical measurements and devices using crystal spheres and cylinders

A single crystal sphere is sufficient to measure the angular distribution of any linear and nonlinear optical property, provided the sphere mounted on a goniometer head is placed at the

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center of a Euler circle as shown in Fig. 5(a). An access to any direction of the studied crystal, is obtained by rotating the sphere on itself using the 3 rotations of the Euler circle [2]. The corresponding angles are read with an accuracy of about 1 minute of arc. A first focusing lens properly placed in front of the sphere as shown in Fig. 5(b), enables any incident beam to propagate inside the sphere collinearly to any of its diameter, and that without any aberration. The beam is tunable since emitted by a tunable nanosecond or picosecond optical parametric device. The key issue is that the sphere strictly rotates around its center. It is possible only when this center is in coincidence with the center of the Euler circle. For this adjustment, we use the two translations of the goniometer head and it is achieved when an incident beam ordinary polarized exhibits no deviation at the exit of the sphere [2,5]. A second focusing lens allows the output beam to be collimated after the sphere for its detection.

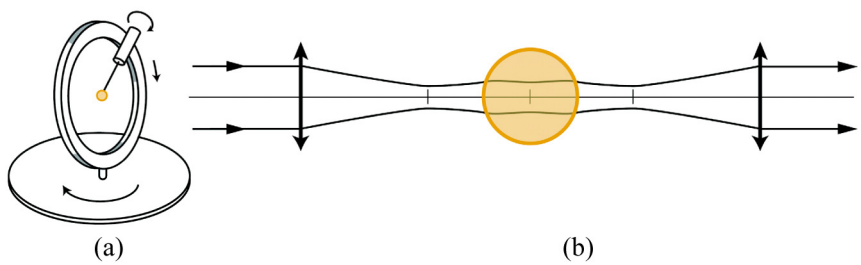

Fig. 5. (a) Sphere placed at the center of a Euler circle. (b) Focusing scheme inside the sphere.

The measurement of the angular distribution of any optical property is usually performed in the dielectric frame (X, Y, Z), in which the real part of the dielectric permittivity tensor is diagonal. This frame is completely linked to the crystallographic frame in crystals belonging to the cubic, hexagonal, tetragonal, trigonal and orthorhombic symmetry groups. However it can rotate in monoclinic or triclinic crystals as a function of any dispersive parameter. Using a tunable optical parametric device, we determine the orientation of the dielectric frame as a function of wavelength by following the corresponding variation of the angle between the two optical axes of the studied monoclinic or triclinic crystal. It was possible by measuring the angular positions of the internal conical refraction effect at the exit of the sphere [2,5]. The accuracy of such a measurement is better than ± 0.5°.

Using the sphere method, we showed to the first time that the dielectric frame is not the proper frame of the imaginary part of the dielectric permittivity [6]. This means that another frame diagonalizes the corresponding matrix, namely (X_{abs}, Y_{abs}, Z_{abs}) for the absorption and (X_{emi}, Y_{emi}, Z_{emi}) for the spontaneous or stimulated emission [6]. The orientation of these frames correspond to symmetry axes of the angular distribution of absorption or emission measured in polarized light, as shown in Fig. 6(a) [6]. When this distribution was measured as a function of wavelength, the symmetry axes rotation revealed an orientation of the corresponding frame strongly depending on the considered electronic transition [6].

The sphere method brought also a lot in the frame work of nonlinear optics since it enabled to measure directly with an accuracy of ± 0.5° the phase-matching angles of any frequency conversion process: Second- or Third-Harmonic Generations (SHG or THG) with one tunable input beam [3, 4], Sum- or Difference- Frequency Generation (SFG or DFG) with two input beams combined collinearly inside the sphere. Given a wavelength and polarization scheme, a phase-matching direction is detected when the conversion efficiency reaches a maximum as a function of the angle. An example of the corresponding recorded phase-matching curve is given in Fig. 6(b) [11]. It can be used *per se* for the design of parametric devices using the studied crystal. But it can also enable to determine the Sellmeier equations valid in the full transparency range of the crystal by fitting simultaneously several tuning curves recorded in the same sphere [2,3, 7–11]. Furthermore, the sphere is the only way to measure the conversion efficiency along a tuning curve, which allows us to determine both the magnitudes and relative signs of the coefficients of the second- or third-order electric susceptibility tensors [3].

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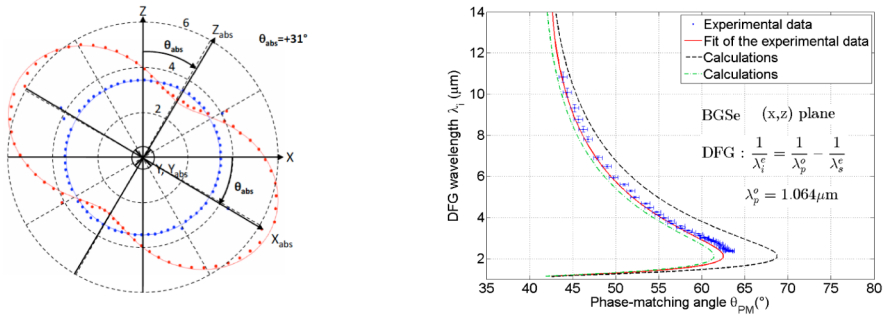
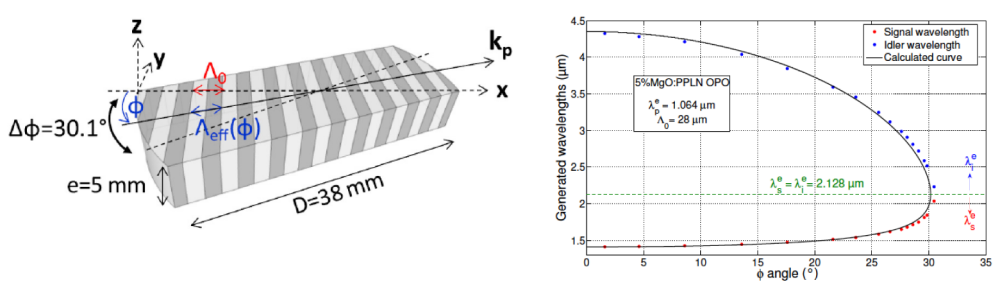


(a) (b)

Fig. 6. Angular distribution recorded in polarized light of: (a) the absorption at 812 nm through a 7.44-mm-diameter Nd^{3+}:YCOB sphere [6]; (b) the DFG tuning curve using a 7.55-mm-diameter BGSe sphere [11].

Cylinders provide widely and continuously tunable optical parametric devices. As an example, we designed Optical parametric Oscillators (OPOs) by inserting a full or partial cylinder between two plane mirrors [12,13] while pumped by a monochromatic wavelength. The rotation axis of the cylinder was chosen to be orthogonal to the plane containing the phase-matching (PM) or quasi-phase-matching (QPM) tuning curve of interest. It is then possible to access any PM or QPM direction located in the cylinder plane, and to generate the corresponding phase-mathing wavelengths without changing Fresnel losses or any spatial distortion, and with less non-colinearity at the level of the wave vectors. The cylinder can be partial as the 5%MgO:PPLN one shown in Fig. 7(a). It was shaped to access only the phase-matching directions for the wide and continuous spectral tuning OPO emission shown in Fig. 7(b). We found energetic performances similar to those of OPOs based on slabs [12–14]. The main advantage of such a shape compared with a multi-grating slab is that the tunability is perfectly continuous. Furthermore, the wavelength over the transverse section of the beam is more homogeneous than in the case of fan-shaped gratings.



(a) (b)

Fig. 7. QPM 5%MgO:PPLN (Λ_0 = 28 µm): (a) partial cylinder, (b) corresponding OPO tuning curve [13].

6. Conclusion

The techniques of shaping spheres and cylinders that we delopped are now mature. They enable the most accurate measurements of the angular distributions of the absorption, laser emission, or phase-matching conditions, as well as the implementation of widely and continuously tunable OPOs. Next we will perform these measurements as a function of temperature between – 40°C and + 40°C, to take advantage of this new dispersive parameter.