Čerenkov second harmonic microscopy for three-dimensional ferroelectric domain visualization

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Abstract: we show that when a laser beam is tightly focused on a single boundary between two 180° anti-parallel ferroelectric domains, a strong enhancement of the Čerenkov second-harmonic generation in the direction perpendicular to the boundary occurs. Using this effect we develop a scanning Čerenkov second-harmonic microscopy which allows direct, three-dimensional and non-destructive imaging of the inverted domain pattern with sub-micrometer resolution.

Engineered ferroelectric domain structures, also known as $\chi^{(2)}$ nonlinear photonic crystals (NPC) or optical supperlattices, are commonly used in nonlinear optics for frequency conversion of laser radiation [1]. The resulting one- or two-dimensional spatial modulation to the sign of the nonlinearity $\chi^{(2)}$ in NPC enables one to quasi-phase match frequency sum or difference mixing or even Čerenkov-type second-harmonic generation (SHG) [2]. The Cerenkov frequency doubling represents the noncollinear SHG where the second-harmonic (SH) is generated at an angle with respect to the propagation direction of the fundamental wave, for which the longitudinal phase-matching condition $k_2\cos\theta=2k_1$ is fulfilled. Here k_2, $2k_1$ denote the wave vectors of the fundamental and SH waves, respectively, and θ is the angle between these two vectors [see Fig. 1 (a)].

Nowadays a variety of ferroelectric domain structures are almost routinely fabricated in crystals such as $LiNbO_3$, $LiTaO_3$ and $KTiOPO_4$, by the electrical-field poling technique. The direct visualization of such inverted domains inside the materials, however, still remains a major challenge. The traditional techniques for domain imaging, e.g. selective etching and scanning electron/force microscopy can only provide information on the surface structure [3]. The internal structure of NPC, which can be diverse and complex, remains inaccessible for non-destructive observations.

In this contribution, we present a new simple method for high-resolution three-dimensional (3D) imaging of the inverted domains, based on Čerenkov-type SH laser scanning microscopy. We apply this method to study the evolution of the inverted domain structures from the surface into the material depth in several NPC fabricated in different ferroelectric materials.

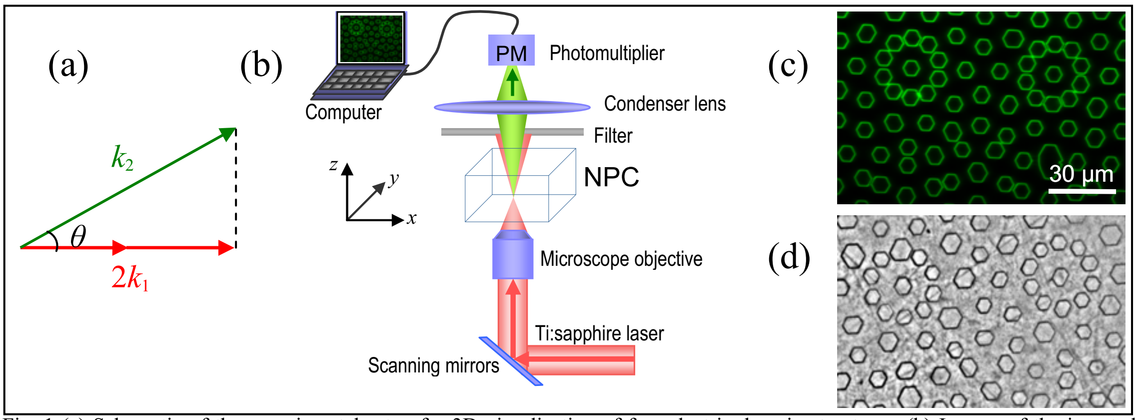

Fig. 1 (a) Schematic of the experimental setup for 3D visualization of ferroelectric domain structures. (b) Images of the inverted domain pattern of a 2D quasi-periodic NPC obtained via laser scanning Čerenkov SHG microscopy inside the NPC; (c) The similar domain structure obtained via optical microscopy after selective etching of the NPC surface. (d) SHG patterns (blue spots) imaged with a CCD camera when the fundamental beam was focused on the corresponding domain boundary of the $LiNbO_3$ NPC.

The experimental setup, shown in Fig. 1 (b), consists of a femto-second laser coupled to a laser scanning microscope and tightly focused into the NPC. The NPC can be scanned in three dimensions, and the generated SH signal is detected by a photomultiplier and fed into a computer. Figure 1 (c) shows a typical *x-y* scan of the SHG

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intensity from a 2D quasi-periodic NPC formed in congruent $LiNbO_3$ crystal with the focal plane located at a depth of ~10 µm above the polished lower surface of the sample. Bright SH intensity patterns in a form of hexagons can be clearly seen. These hexagons represent boundaries of the inverted domains as evident from the comparison with the conventional optical microscopy image [Fig. 1 (d)] obtained from the surface of similar NPC after selective chemical etching. Figure 1 (c) shows that the generated SH signal is strongly enhanced when the beam focal spot is positioned on a boundary between the inverted and non-inverted parts of the NPC. In order to understand the origin of this enhancement we have modified our experimental setup, replacing the condenser lens and the photo-multi-player with a diffusive screen and a color CCD camera. When we placed the focus spot of the fundamental wave on the domain walls in the sample, we observed two SH beams emitted non-collinearly with respect to the fundamental beam. The measured external angle between the SH and fundamental beams was 52.4^o, in good agreement with the theoretical value for the Čerenkov angle of 51.0^o derived on the basis of longitudinal phase matching condition.

In order to explore its universality, we applied the Cerenkov SHG microscopy to examine the inverted domains in NPC made of different ferroelectric crystals, as shown in the top row of Fig, 2. In all cases the Čerenkov-type SHG laser scanning microscopy works nicely, producing high contrast images. Furthermore, the method offers an exceptional spatial resolution, which is below the diffraction limit for the excitation laser wavelength of 820 nm. As shown in Fig. 2(e) even domain boundaries separated by less than 250 nm can be easily resolved.

The Čerenkov-type SHG laser scanning microscopy can also produce 3D images by combining *x-y* scans measured at different depth *z* inside the NPC. The bottom row of Fig. 2 shows such 3D images of the inverted domain distribution hidden inside the short-range ordered NPS, from which one can determine the quality of poling in the bulk of the crystal. Particularly, the processes of domain deformation and domain merging are visualized in Fig. 2 (h) and (i), respectively.

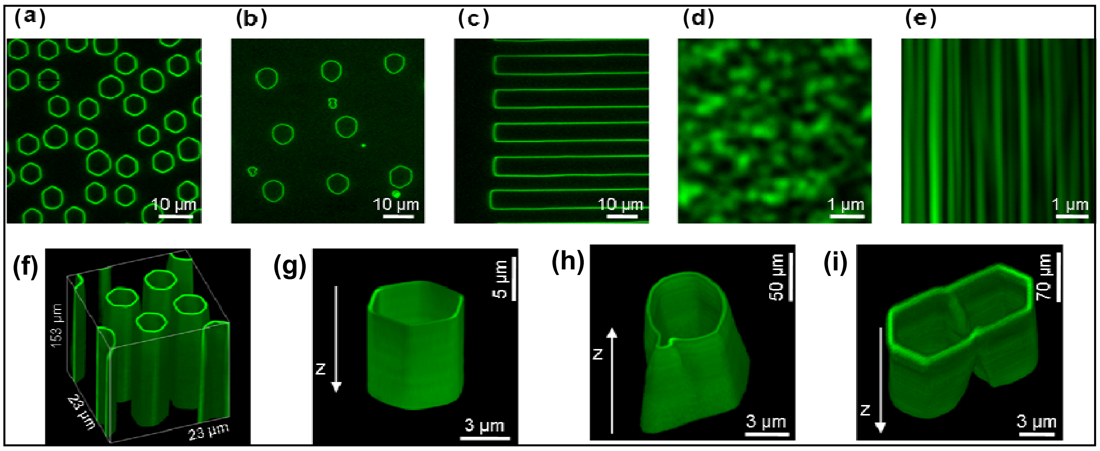

Fig. 2 Top row: Ferroelectric domain structures imaged by the Čerenkov-type SHG microscopy, taken with the focal plane of the fundamental beam located between 10 to 20 µm inside the corresponding NPC: (a) Congruent $LiNbO_3$ NPS with 2D short-range ordered domain structure. (b) Stoichiometric $LiTaO_3$ NPS with 2D quasi-periodic domain structure. (c) $KTiOPO_4$ NPS with 1D periodic domain structure. (d) and (e) As-grown $SrBaNbO_3$ crystal with naturally random domain structure at the *z* surface and the *x* surface, respectively. Bottom row: 3D visualization of inverted ferroelectric domains inside congruent $LiNbO_3$ crystal by Čerenkov-type SH scanning microscopy.

In conclusion, we have developed a new method for direct 3D imaging of the ferroelectric domains structures with sub-diffraction limit resolution. The method is based on the strong enhancement of the Čerenkov SHG at the domain wall regions. We apply this new technique to visualize the geometry of the inverted domains in the bulk of the medium, showing details such as, for instance, the deformation of the domain shapes or the merging of the individual domains into larger entity. The ability to extract such information is important for a better understanding of domain reversal process and for the control of the domain structures. The Čerenkov SHG microscopy provides a powerful tool that will further inspire the design and development of new and sophisticated domain structures for advanced photonic applications.

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