Second-harmonic imaging from a modulated domain structure

Yong Zhang,1,2 Fuming Wang,1,3 Katrina Geren,2 S. N. Zhu,1 and Min Xiao1,2,*

1National Laboratory of Solid State Microstructures and Department of Physics, Nanjing University, Nanjing 210093, China
2Department of Physics, University of Arkansas, Fayetteville, Arkansas 72701, USA
3Current address: Geballe Laboratory for Advanced Materials, Stanford University, Stanford, California 94305, USA
*Corresponding author: mxiao@uark.edu

We present a new second-harmonic (SH) imaging technique to study the domains in a hexagonally poled LiTaO₃ nonlinear photonic crystal by using a femtosecond laser. By detecting the SH images at different planes, the distribution of the 180°-inverted ferroelectric domains can be characterized, and the contributions of different nonlinear tensor components, modulated differently in the domain structure, can be selectively determined. Fundamental understanding and potential applications of such SH imaging techniques for the inverted nonlinear domain structures are presented and discussed. © 2010 Optical Society of America

OCIS codes: 190.2620, 140.3515, 160.1245, 190.4720.

Periodically inverted ferroelectric domain structures have been fabricated in recent years for broad applications in efficient nonlinear wavelength conversions, optical switching, and information storage [1,2]. Two-dimensional domain structures, especially with hexagonal symmetry, can provide an increased range of tunability for the second-harmonic generation (SHG) owing to the increase of reciprocal lattice vectors [3]. Also, the two-dimensional domain structures can produce noncollinear second-harmonic (SH) waves caused by a noncollinear reciprocal vector, scattering, or Cerenkov radiation [4–6]. In Cerenkov SHG, the SH waves are emitted at the Cerenkov angle \( \theta_c = \arccos \left( \frac{\nu'}{\nu} \right) \), where \( \nu' \) and \( \nu \) are the phase velocities of the SH wave and the fundamental wave, respectively. Cerenkov SHG occurs as \( \nu' > \nu \).

Direct observations of ferroelectric domains are routinely performed by using linear optical polarization microscopy. A SHG technique has been recently developed to visualize various ferroelectric, ferromagnetic, and multiferroic domain structures [7,8] that relies on the interference between the SH waves generated from various domains. SH radiation patterns in a nonlinear crystal normally involve several nonlinear tensor components. Here we show that, by focusing the femtosecond laser beam on different spot sizes of a two-dimensionally inverted LiTaO₃ slice, the contributions from different nonlinear tensor components can be better determined and studied from the near-field and far-field SH patterns.

The experimental studies are done with a z-cut hexagonally poled LiTaO₃ (HPLT) [Fig. 1(a)] slice of 0.5 mm thickness. The interval between the inverted domains (center to center) is 9.0 μm. When the domain structure is fabricated, the direction of the spontaneous polarization of the inverted (negative) domains is sequentially inverted [1]. In addition, the domain inversion process is also accompanied by lattice distortions across domain walls [9], which could also modify the generated SH radiation patterns.

The excitation source is an 800 nm mode-locked Ti:sapphire femtosecond laser. As shown in Fig. 1, the focused laser beam propagates along the z axis of the crystal with its polarization parallel to the x axis of the crystal. The generated SH signals are either collected by a camera for near-field imaging [Fig. 1(b)] or projected onto a screen 100 mm away from the sample [Fig. 1(c)]. The fundamental beam is filtered out by a bandpass filter. These SH radiation patterns reflect contributions from different nonlinear tensor components. Although the far-field images [Fig. 1(c)] can clearly show separate collinear and noncollinear radiation patterns, the near-field images are more complicated, depending on the size of the focal spot on the sample relative to the domain structures, as will be discussed later.

Before using HPLT, we first measure the SH patterns of an unpoled LiTaO₃ as a reference. A lens of \( f=200 \text{ mm} \) is first used, which gives a focal spot of \( \sim 50 \text{ μm} \) diameter on the sample. To see the details of the SH images, a lower laser power (100 mW) is used. The far-field SH pattern is shown in Fig. 2(a), which includes a bright spot at the center and a uniform ring pattern. The central spot is obviously from

Fig. 1. (Color online) Experimental setup: (a) sample domain structure, (b) near-field SH imaging from the camera, (c) far-field SH imaging projected on the screen.
the collinear SHG. The ring is from the Cerenkov SHG. The Cerenkov angle is given by \( \theta_c = \arccos(\nu/\nu') = \arccos(2k_w/k_{2w}) \). Here, \( k_w = 16.91 \, \mu m^{-1} \) and \( k_{2w} = 35.86 \, \mu m^{-1} \) are the fundamental (800 nm) and SH (400 nm) wave vectors, respectively. Considering the refraction at the crystal surface, the calculated Cerenkov angle is 49.5°, which is very close to the measured 50.0°. With the fundamental beam polarization parallel to the \( x \) axis, only \( d_{31} \) and \( d_{21} \) in the nonlinear tensor could be involved in the SHG [10]. Since the fundamental beam propagates along the \( z \) axis, only \( d_{21} \) contributes to the collinear SHG whereas the noncollinear ring is generated by both \( d_{21} \) and \( d_{31} \) [11–14].

Next, the HPLT [Fig. 1(a)] is used with the same experimental conditions. The focal spot (50 \( \mu m \)) covers more than one unit cell of the domain structure, which includes seven inverted domain spots [Fig. 2(g)]. The far-field SH pattern has a structure with a hexagonal symmetry both in the central spot and in the outer ring [Fig. 2(b)]. By considering the polarization and propagation directions of the fundamental beam and the domain structure, \( d_{21} \) contributes mainly to the collinear central spot [see Fig. 2(a); the ring is much weaker than the central spot], and both \( d_{21} \) and \( d_{31} \) have contributions to the SH ring [11–14]. Actually, the domain walls can contribute additional nonlinearity in the \( x-y \) plane, as will be discussed later [15]. The reason for having a weaker collinear central spot, in comparison with the uninverted sample [Fig. 2(a)], is the involvement of domain structures, which can greatly enhance the Cerenkov SHG [11,12]. Figure 2(e) depicts the near-field image.

Although both \( d_{21} \) and \( d_{31} \) contribute to this SH radiation pattern, since the diffractions of the SH waves have not fully developed at such a short propagation distance, the central part of the image is still mainly from the SH process involving \( d_{21} \), because \( d_{21} > d_{31} \) (\( d_{21} = 1.6d_{31} \) [16]) and \( d_{21} \) dominates the SH process in the forward collinear direction. It is interesting to note that the orientations of the hexagonal symmetries of the central patterns in Figs. 2(e) and 2(b) are different with a rotation of 30°. The near-field image [Fig. 2(e)] has the same orientation as the domain structure itself [Fig. 2(g)].

When an \( f=75 \) mm lens is used, the laser spot on the sample is reduced to 20 \( \mu m \), which basically covers only one unit cell. In this case, although the far-field SH pattern [Fig. 2(c)] is almost the same as the previous one [Fig. 2(b)], the near-field SH image [Fig. 2(f)] looks different from the one in Fig. 2(e). Within the beam spot (<20 \( \mu m \) in the center), there are hexagonal symmetric spots that are rotated from the symmetry of Fig. 2(e). In the center (<10 \( \mu m \) diameter), the SH signals are mainly from the collinear SHG caused by \( d_{21} \), and the scattered SH waves outside the central area (>10 \( \mu m \)) are caused by the noncollinear SHG due to \( d_{31} \).

To understand the observed SH patterns, numerical simulations are made to consider contributions from different nonlinear tensor components. Since the far-field patterns [Figs. 2(b) and 2(c)] are easily understood from calculating the collinear SHG and the Cerenkov SHG, we concentrate only on the near-field SH patterns [Figs. 2(e) and 2(f)]. Since \( d_{31} \) and \( d_{31} \) contribute quite differently to the SH patterns, we can consider them separately by using different models. From our experimental results and simulations, the effective transverse nonlinear coefficient \( d_T \) in our HPLT sample, which consists of the \( d_{21} \) from the LiTaO\(_3\) crystal and the nonlinearity from the domain walls [15], seems to have the same sign in both positive and negative domains. However, \( d_T \) is distorted at domain walls [9]; so is the refractive index, which results in a scattering of the nonlinear signals at the domain walls. In this case, each domain wall can be simplified as a radiation light source. If we consider the excitation spot of 20 \( \mu m \) to cover just one unit cell, the \( d_T \) contributions can be modeled as due to seven light sources (labeled 0–6 in Fig. 2(g)), each of which is a domain spot surrounded by a domain wall. For simplicity, the background domain contribution is neglected here. The image plane in the simulation is set at an effective distance of \( L_{eff}=5 \lambda \) (\( \lambda=400 \) nm). The seven light sources are in phase, so at the image plane, the interference gives sharp spots with hexagonal symmetry similar to the observed SH image pattern of Fig. 2(e). We can conclude that the central pattern in Fig. 2(e) is caused mainly by \( d_T \) for collinear SHG. Although previous reports indicate that \( d_{31} \) is reversed in periodically poled lithium niobate [17,18], the nonlinear coefficients in the \( x-y \) plane are more complicated when the fundamental laser beam propagates along the \( z \) axis of the crystal. In this case, the domain structure (walls) can gener-
ate extra nonlinearity in the \(x-y\) plane \([15]\), so we consider an effective nonlinear coefficient \(d_7\) in the \(x-y\) plane, which is composed of the \(d_{21}\) from the LiTaO\(_3\) crystal and the nonlinearity from the domain structures (walls). For \(d_{31}\), the model is more complicated, since the sign of \(d_{31}\) is changed in the inverted domains. In this case, we have to consider both the inverted (inside the domain walls) and the uninverted (outside the domain walls) domains. To simplify the problem, other than the seven inverted domains with \(-d_{31}\) (spots 0–6), we add six additional spots with \(+d_{31}\) (spots 7–12 in Fig. 2(g)), to take into account the contributions from the uninverted background. In the simulation, these domain spots act as light sources with an opposite phase (due to the different signs of \(d_{31}\)). At \(L_{\text{eff}}=5\lambda\), the simulated SH pattern has an interesting hexagonal symmetry [Fig. 2(i)]. The large diffraction pattern looks similar to the observed pattern shown in Fig. 2(f), except at the center (<10 \(\mu\)m, where \(d_{21}\) is dominant due to the collinear SHG), which indicates that the \(d_{31}\) component has significant contributions at the near-field image when the fundamental beam is focused to be smaller to cover just one unit cell. In this case, the pattern area from \(d_{31}\) is much larger than that from \(d_{21}\). It is interesting to note that the hexagonally symmetric center spots in Figs. 2(b) and 2(c), which are due mainly to the collinear SHG caused by \(d_{21}\), are rotated by 30° relative to the hexagonal symmetry in Figs. 2(e) and 2(h). Our simulation with the \(d_{21}\) SH process [inset in Fig. 2(h)] shows that, at \(L_{\text{eff}}=5\lambda\), the hexagonal symmetric pattern evolves to the same orientation as the far-field one, which confirms our understanding of the SH radiation in the domain structure.

When the focal spot is further reduced to approximately 3 \(\mu\)m by a lens \(f=5\) mm, it can only excite a pair of inverted–uninverted domain spots. Therefore, only one pair of the Cerenkov segments exists at a given excitation position in the far-field screen (Fig. 3), corresponding to different domain pairs. Here, another nonlinear component \(d_{33}\) can no longer be ignored as the fundamental beam is tightly focused. As for \(d_{31}\), the sign of \(d_{33}\) is also flipped in the inverted domains, so we can simulate the SH radiation patterns using the same model as \(d_{31}\). The simulations show that Figs. 3(a)–3(c) correspond to the excited light-source pairs of 0&8, 0&9, and 0&10, respectively. Because of the large divergence angle of the fundamental beam, the observed segments are thick and the central spot is too weak to be seen.

In summary, we have investigated the far-field and near-field SH images from a hexagonally poled LiTaO\(_3\). By varying the focal spot size on the sample, different contributions from various nonlinear susceptibility components are identified. Simulations with simplified models have been developed and used for comparison with the observed SH patterns, which have greatly enhanced our ability to understand various nonlinear SH processes and their contributions to the spatial patterns.

This research is supported by the Arkansas Institute for Nanoscale Materials Science and Engineering and by the National Natural Science Foundation of China, 10534020.

References