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SINGLE DOMAIN WALL EFFECT ON PARAMETRIC PROCESSES VIA CHERENKOV-TYPE PHASE MATCHING

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We report on a new character of single domain wall (DW) of electrically-poled ferroelectric crystal which can modulate parametric processes via Cherenkov-type phase matching. Experimental result shows that the effective nonlinear polarization is confined in DW, and its phase velocity can be modulated when incident light is off the domain wall's direction. These effects lead to novel Cherenkov second harmonic generation (CSHG), and other modulated parametric process, such as Cherenkov sum frequency generation (CSFG).

Keywords: Domain wall; ferroelectric; CSHG; Cherenkov; parametric process.

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1. Introduction

A charged particle moving faster than light in a medium can drive surrounding atoms to emit coherent light called Cherenkov radiation (CR).[1] CR is always observed in a cone defined by Cherenkov angle $\theta_C = \arccos(v'/v)$, where v is the velocity of the moving charged particle and v' is the phase velocity of CR. It can be seen that CR occurs only when $v > v'$. A similar phenomenon called Cherenkov second harmonic generation (CSHG) is observed in nonlinear optics processes,[2–4] when the phase velocity of nonlinear polarization propagates faster than the phase velocity of free light of second harmonics in medium. While these two processes are similar, CSHG has unique features. For example, there is no charged particle but a spatially extended nonlinear polarization driven by the incident light field. Typical setup for CSHG is waveguide structure with nonlinear substrate.[3] In recent years, CSHG has also been reported in SBN crystal.[5,6] and periodically poled ferroelectrics,[7,8] for which samples have domain walls (DW) in them. There are new features of DW among those reports, such as optical birefringence, strain, local electric fields and electromechanical contrast, and so on.[9]

In this letter, we study parametric processes via Cherenkov-type phase matching in a single DW theoretically and experimentally. We find that the nonlinear polarization, which generates the observable CSHG, is confined in DW. By changing the angle of incident light with respect to DW, we can easily change the phase velocity of the nonlinear polarization and consequently change the Cherenkov angle of the CSHG. As further evidence, Cherenkov sum frequency generation (CSFG) modulated by DW is also demonstrated in our experiment.

2. Experimental Method

CSHG has been reported in periodically poled ferroelectrics with multiple DWs.[7,8] In order to investigate the influence of DW on the CSHG, we focused our study on CSHG in single DW. Single DW with high quality is fabricated in 1 mm Z-cut $LiNbO_3$ sample as shown in Fig. 1. A Y-polarized incident light of 100 fs pulses centered at 800 nm with average power of 200 mW is loosely focused ($f = 15\,\text{cm}$) in the DW along z direction with spot size about 100 μm. It can be seen from the screen as shown in Fig. 1 that a pair of well-collimated CSH has been generated, and they are symmetrical to the DW. In comparison, we cannot observe any CSHG in bulk $LiNbO_3$ with pure domain. This experiment demonstrates that CSHG in ferroelectrics indeed originates in their DW, but not periodically poled structures.

3. Discussion

A more important phenomenon in this experiment is a well-collimated CSH pair which is symmetrical to the DW. Let us consider the situations of CSHG in bulk ferroelectrics and planar waveguide. CSH generated in bulk ferroelectrics is conical,[2] because the stimulated nonlinear polarization is determined by the incident beam of cylindrical symmetry without confinement of material shape. Relatively, in the case of planar waveguide, because of the restrictions of refractive index difference,

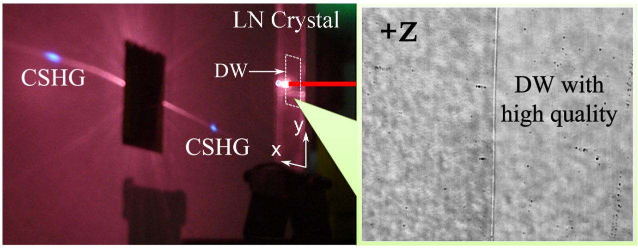
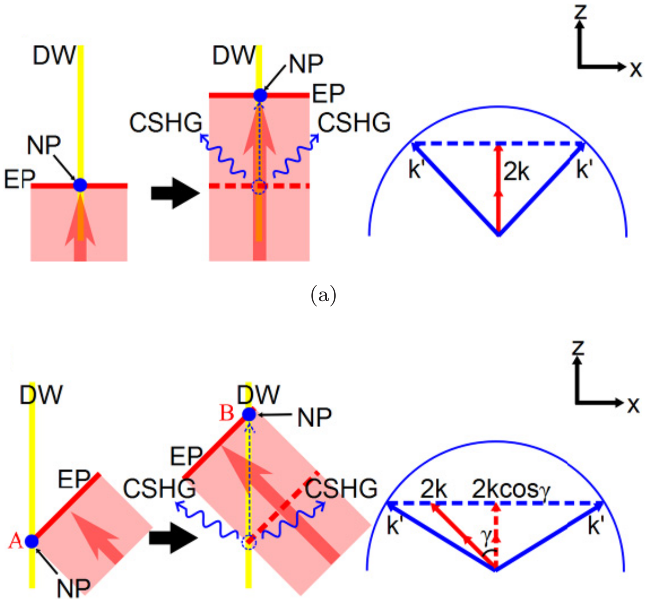


Fig. 1. Pattern of CSH generated in DW. In the left part, the red line represents the incident light and the dashed area is a single DW sample fabricated by poling technique. LN means lithium niobate. The right picture is the DW observed in microscopy.

the fundamental beam is trapped in the waveguide, and only the medium within the waveguide is stimulated, which means the geometry of the nonlinear polarization material is planar. When the phase velocity of the fundamental beam v in the waveguide is larger than the phase velocity of second harmonic (SH) v' in the substrate, the harmonic radiation emits into the substrate at Cherenkov angle.[2–4] We can see that the pattern of CSHG is determined by the geometry of the stimulated nonlinear polarization material. In our experiment of CSHG at domain wall, the well-collimated CSH indicates only the planar domain wall to be the only stimulated nonlinear polarization material, the typical width of which is believed to be less than 100 nm in ferroelectrics.[10] Although the incident beam waist of about 60 μm, nonlinear polarization only occurs in the domain wall area.

Energy and momentum conservation are both satisfied in this CSHG process in DW. Energy conservation is fulfilled by converting two photons with frequency ω to one photon with frequency 2ω. Momentum conservation is fulfilled by Cherenkov-type phase matching condition, to be discussed in detail as follows. See Fig. 2(a), when incident light is along the direction of DW, it drives second order nonlinear polarization propagating along DW at the same phase velocity $v_{\mathrm{np}} = v$. Free SH propagates at a smaller phase velocity v' in the same medium. Cherenkov angle is



(a)

(b)

Fig. 2. Schematic depiction of CSHG processes and the corresponding phase matching condition when (a) the incident light is along the direction of DW; and (b) the incident light has an angle with respect to the DW in x–z plane. EP and NP mean equiphase surface and nonlinear polarization respectively. The red area and the bold large arrow represent the incident beam and its direction.

determined by $\theta_C = \arccos(v'/v)$. We transform v and v' into momentum space and obtain the Cherenkov-type phase matching conditions,

$$\cos\theta_C = \frac{2|\vec{k}|}{|\vec{k}'|} \tag{1}$$

where $\vec{k}$ and $\vec{k}'$ are the wave vectors of incident light and SH in the medium respectively. The triangle relationship of these two wave vectors is also shown in Fig. 2(a). This is typical Cherenkov-type phase matching. However, since the nonlinear polarization is confined in DW, it is easy to find that new interesting CSH can be generated. As shown in Fig. 2(b), when we rotate the incident light in the X–Z plane so that there is an angle γ with respect to the Z-axis, the forced nonlinear polarization in DW will not propagate collinearly with the incident light. At the beginning, the equiphase surface of incident light drives nonlinear polarization in DW at point A, then with the propagation of this equiphase surface at phase velocity of v, the nonlinear polarization at point B in DW is stimulated after a period of time Δt. The distance that nonlinear polarization propagates is from point A to point B, which is equal to $v\Delta t/\cos\gamma$. We can see that the phase velocity of the nonlinear polarization has been increased to be $v_{\mathrm{np}} = v/\cos\gamma$. Free SH still propagates at phase velocity v', so the Cherenkov angle becomes $\theta_C = \arccos(v'\cos\gamma/v)$. We can transform this relationship into momentum space again and get new phase matching condition for incident light with arbitrary incident angle γ with respect to DW in X–Z plane,

$$\cos\theta_C = \frac{2|\vec{k}|\cos\gamma}{|\vec{k}'|}. \tag{2}$$

Experimentally, we rotate the sample with single DW in X–Z plane and obtain the result that the CSHG angle increases with the incident angle, as shown in Fig. 3. The measured external CSHG angle θ_c changes with different incident angle γ,

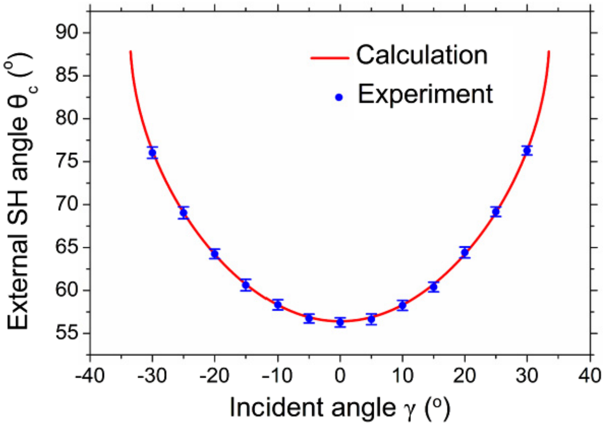


Fig. 3. External Cherenkov angle of CSHG changes with the incident angle.

which is in good agreement with the theoretical calculations in Eq. (2). By changing the incident angle, it is easy to control the phase velocity of the nonlinear polarization from v (the velocity of incident beam) to infinity. The CSHG can be modulated continuously. From our experiments, we can conclude that single DW plays a key role in this process.

As discussed earlier, CSH can be generated in any normal dispersive nonlinear medium theoretically. Then why is it enhanced in DW? From Maxwell's equations, we derive the inhomogeneous vector wave equation for CSHG

$$\nabla \times \nabla \times \vec{E} - \frac{\omega_h^2}{c^2}\varepsilon \vec{E} = \omega_h^2 \vec{P} \tag{3}$$

where ω_h is the frequency of the SH, ε is the dielectric constant, $\vec{P}$ is the second order nonlinear polarization. Detailed solving process is discussed in the previous literature,[11] and the solution for electric field of CSH is

$$\vec{E}(r) = \frac{1}{r}\frac{\omega_h^2}{4\pi\varepsilon_0 c^2}\vec{P}VX(n,n')D(n,n')e^{-i\frac{\omega_h}{c}n'r} \tag{4}$$

where n and n' are the refractive indices of incident light and CSH respectively. $X(n,n')$, and $D(n,n')$ are functions of n and n', and they equal to unity in the limit of a small source, and V is also a constant. From Eq. (4), we find that the only reason for the enhancement of CSHG in DW is the enhancement of $\vec{P}$. Since $\vec{P} = d_{\text{eff}}\vec{E}\vec{E}$, d_{eff} in DW must have been enhanced. In our previous study,[12] the CSHG efficiency in PPLN is measured to be about 1%, which is several orders of magnitude greater than that of 10^{-10} in bulk LiNbO$_3$.[2] Eliminating the influence of the number of domain walls in PPLN, we estimate the enhancement of d_{eff} in DW to be about 10^3. Consequently, enhanced effective nonlinear coefficient leads to enhanced nonlinear polarization confined in DW, which emits observable CSHG.

A more complicated parametric process via Cherenkov-type phase matching is available. When two incident light beams at the frequency of ω_1 and ω_2 are injected at angles γ_1 and γ_2 with respect to DW in the X–Z plane, each of them can generate a pair of CSHs at the angles satisfying Eq. (2). When they exactly overlap in DW, as shown in Fig. 4, each of them can force a nonlinear polarization with wave vectors of $\vec{k}_1$ and $\vec{k}_2$, and the sum frequency polarization is created by

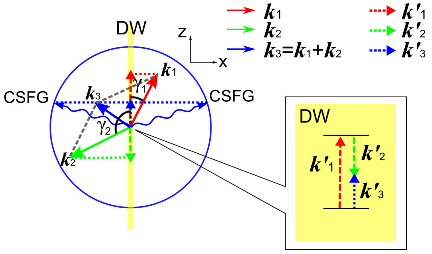


Fig. 4. Schematic of modulated Cherenkov SFG in DW. The two incident lights can force a sum frequency nonlinear polarization which radiates the observed Cherenkov SFG.

a combination of these two wave vectors, $\vec{k}_3 = \vec{k}_1 + \vec{k}_2$. When the existence of DW confined the sum frequency polarization, the wave vector has been modulated to be $|\vec{k}_3'| = |\vec{k}_1|\cos\gamma_1 + |\vec{k}_2|\cos\gamma_2$. Then we get the definition of the emitting angle of the Cherenkov SFG, which also satisfy the general Cherenkov phase-matching condition for DW-modulated frequency up-conversion processes,

$$\cos\theta_C = \frac{|\vec{k}_{\omega_1}|\cos\gamma_1 + |\vec{k}_{\omega_2}|\cos\gamma_2}{|\vec{k}_{\omega=\omega_1+\omega_2}|} \tag{5}$$

In this equation, the momentum conservation is fulfilled by Cherenkov-type phase matching condition, and then CSFG can also be modulated by DW. If γ_1 and γ_2 become 0, Eq. (5) becomes the definition of typical CSFG.[13]

Experimentally, we split the incident pulse of the femto-second laser source into two identical pulses and tune the delay of one to make them overlap precisely in the DW at different incident angles [shown in Fig. 5(a)]. Three pairs of SH beams are observed in our experiment. Figure 5 shows one side of the SH pattern. In Fig. 5(b), the outer and the inner SH spots are DW-modulated CSH generated by the two beams of incident light respectively, and the middle one is DW-modulated CSF generated by them together. If we tune the delay of one pulse and cause them not to overlap, the middle one disappears at once [Fig. 5(c)]. We can see in Figs. 5(d) and 5(e), when either of the two incident beams is blocked, the corresponding CSHG and one CSFG disappear. If we rotate the sample, which can change the incident

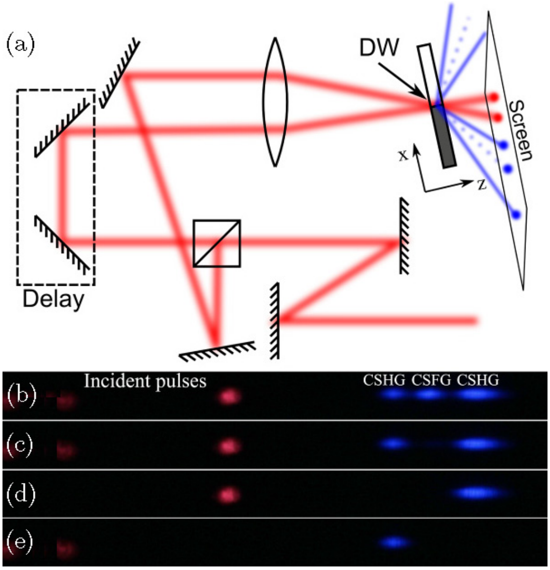


Fig. 5. (a) Schematic of the setup used for the DW-modulated CSFG; (b) shows the observed CSHGs and CSFG when the two incident pulses overlap in DW; (c) is the situation when the two incident pulses do not overlap in DW; (d) and (e) show the results when one of the incident pulses is blocked.

angles of the two pulses simultaneously, the CSH spots and the CSF spot move continuously. When the two incident beams are at the symmetry of the domain wall, namely $\gamma_1 = \gamma_2$, these two CSH and one CSF have the same Cherenkov angle, and the three spots gather together. The existence of DW imposes additional confinement on the propagation of nonlinear polarization without changing the incident pulses, so that it can modulate the nonlinear polarization continuously.

4. Conclusion

In conclusion, we demonstrate the significance of ferroelectric DW on parametric processes via Cherenkov-type phase matching condition theoretically and experimentally. DW-modulated CSHG and CSFG are observed in our experiment, and are consistent with our prediction. The nonlinear polarization forced by incident light is confined in DW, and by solving the Maxwell's equations, we find that the reason is the enhanced effective nonlinear coefficient in DW. This new characteristic of DW should draw greater attention, and more information on the origin of the enhancement of nonlinearity in DW should be obtained from future studies.

Acknowledgments

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