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## Formation of Small Size Polarization Domain Inversion for High-Efficient QPM-SHG Device -Proposal of Full Cover Electrode Method-

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### I. Introduction

Recently, a quasi-phase matched second harmonic generation (QPM-SHG) device using the single crystal of ferroelectric materials, such as LiNbO<sub>3</sub> (LN) and LiTaO<sub>3</sub> (LT), is strongly demanded in the field of opto-electronics [1,2]. The key technology to fabricate an efficient QPM-SHG device is a formation of a fundamental periodic domain inversion, but it is not perfectly fabricated until recently. Up to now, there are many reports of the domain inversion formation [1-3]. Among them, the fabrication of comb electrode by the lift-off process is the most useful method. However, it becomes more difficult as the periodicity is decreased less than 1 $\mu$ m. In this paper, we have proposed a new method named "Full Cover Electrode Method (FCEM)" to fabricate small size periodic electrode without forming the comb electrode (Fig.1) [4]. Instead of a forming a comb electrode, inversion voltage is applied overall surface where the periodic resist pattern is formed. In general, we can fabricate 0.1 $\mu$ m width of resist pattern by using an electron beam (EB) lithography. In the FCEM, only the area directly touched to the electrode is inverted due to a convergence of the electric field. A metal electrode or electrolyte solution is used to apply the voltage. The aim of this study is to show a design theory and an actual usefulness of periodic domain inversion by the FCEM.

### II. Method of Fabricating Microscopic Periodical Domain Inversion ("Full Cover Electrode Method")

Full Cover Electrode (FCE) is composed of an insulating material (EB resist) layer which periodically patterned resist layer is formed on the surface and an electrode covering its full surface. An electrode is composed of a metal electrode or an electrolyte solution. Periodically modulated electric field is generated, when the high voltage is applied between the FCE formed on the +z-face of ferroelectric materials and

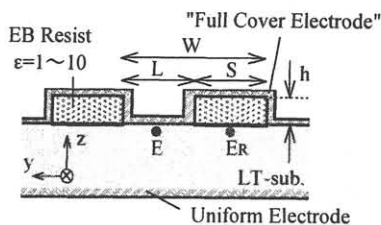


Fig.1 A cross sectional view of "FCEM".

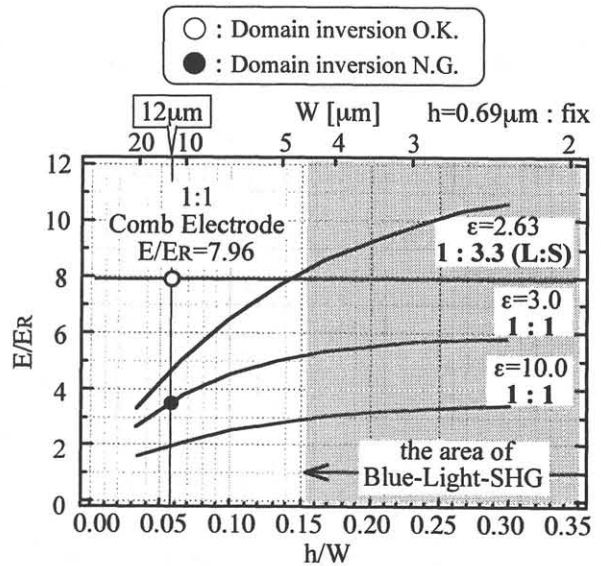


Fig.2 The relationship between E/ER and h/W in LT.

the metal electrode formed on the backside -z-face of the sample. The modulated electric field inverts only the area directly touched to the electrode, because of the convergence of the electric field (refer to Fig.3). The z-component of the electric field distribution in the LT crystal is calculated as a function of width (W), height (h) and dielectric constant ( $\epsilon$ ) of resist by Successive Over Relaxation (SOR) method [5]. Value of the  $\epsilon$  of the resist is changed by condition of formed the electrode, for example, the  $\epsilon$  value of evaporation method and spatter method are about 3 and 10, respectively [4]. The relative dielectric constants of y-component and z-component are  $\epsilon_y=41$  and  $\epsilon_z=43$  for LT, respectively. The intensity of the electric field in the center of the resist and in the center of the electrode are expressed as  $E_R$  and  $E$ , respectively. The relationship between the ratio of h/W and the ratio of E/ER is calculated. Fig.2 shows the relationship between E/ER and h/W in LT, for Line (L) and Space (S) L:S = 1:1 and L:S = 1:3.3. The values of E/ER for the comb electrode for L:S = 1:1 is also shown in the figure. In the FCEM, as the value of h/W becomes larger, the value of E/ER is enhanced. It shows that high value of E/ER is obtained by reducing W or increasing h. High value of E/ER is also obtained by increasing the ratio of S/L, or lowering value of  $\epsilon$ . Moreover, we see from Fig.2 that the value of E/ER at

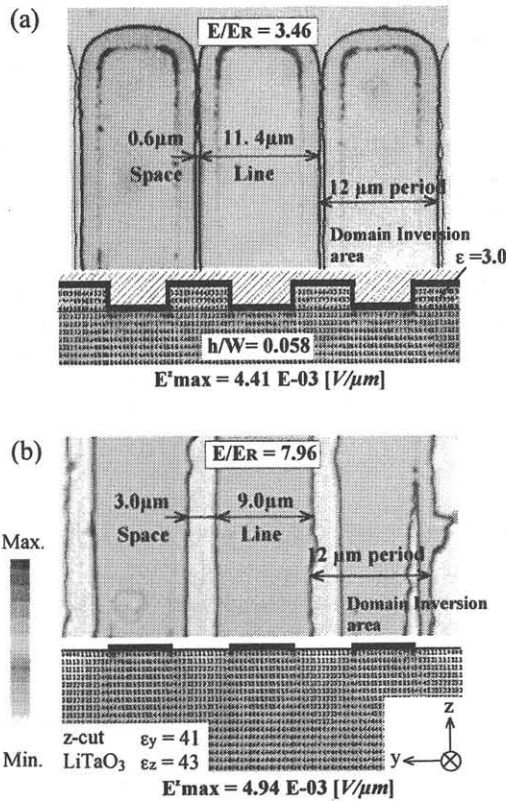


Fig.3 An optical microscope image of the domain inversion in the LT crystal (a) the FCME, (b) comb electrode formed with the lift-off process and the calculated z-component of the electric field in the LT crystal. To reveal inverted area the +z surface is etched.

L:S=1:1 can't rise above the value of  $E/ER$  of the lift-off electrode, however in case of modulated L:S = 1:3.3, the value of  $E/ER$  of the FCME can rise above the value of  $E/ER$  of the lift-off electrode, when the periodic size is reducing. Namely, if domain inversion shape depended on high value of the  $E/ER$  on crystal surface, the FCME would be powerful technique for a narrow period. We examined if domain inversion is bound up with magnitude of the value of  $E/ER$ . In concrete terms, we examined at the points of different value of the  $E/ER$  with the same period.

### III. Experiments

The z-cut LT crystal is used for the substrate. The thickness of the crystal is  $500 \mu\text{m}$ . The resist pattern is formed on +z surface by EB lithography. The thickness of the resist is  $0.69 \mu\text{m}$ . The electrodes are formed on +z surface by the lift-off process or the FCE (Al electrode) over the resist pattern. The uniform electrode on the -z surface are formed by a sputtering method. The high voltage pulse for domain inversion was applied at room temperature. The applied high voltage is  $10.6 \text{ kV}$ , because value of the domain inversion threshold of the sample is  $20.2 \text{ kV}/\text{mm}$ . The pulse width is changed from  $100 \mu\text{s}$  to  $1 \text{ ms}$ . The charge transfer to the crystal is monitored for the detection of domain inversion. After the inversion, the electrodes were removed by mixture of hydrochloric acid and nitric acid.

The inversion pattern is revealed by wet etching using a 1:2 mixture of  $\text{HNO}_3$  and  $\text{HF}$  for LT [6]. The etching pattern is observed by using an optical microscope. Fig.3(a) (b) is shown that optical microscope image of the domain inversion in the LT crystal with  $12 \mu\text{m}$  periodic comb electrode formed with the lift-off process and the FCME with calculated z-component of the electric field in the LT crystal. Both the width and spacing of the electrode are  $6 \mu\text{m}$ . When the value of  $E/ER = 3.46$ , the width of the inverted and non-inverted area were  $11.4 \mu\text{m}$  and  $0.6 \mu\text{m}$ , respectively. On the other hand, when the value of  $E/ER = 7.96$ , the width of the inverted and non-inverted area were  $9.0 \mu\text{m}$  and  $3.0 \mu\text{m}$ , respectively. The results were recognized strong correlation between the value of  $E/ER$  and the domain inversion shape qualitatively. Namely, domain inversion area got in touch with adjacent domain inversion area, in the case of low value of  $E/ER$ . In contrast, non-domain inversion area was appeared, in the case of high the value of  $E/ER$ . In the case of increasing the ratio  $h/W$ , these results seem a beneficial effect of the FCME.

### IV. Conclusions

In this paper, a new method named "Full Cover Electrode Method (FCME)" is proposed for domain inversion of ferroelectric materials, which is most important process for realizing high-efficient QPM-SHG device. We also report the design theory and the actual usefulness of the FCME. The validity and limitation of the method is examined by the SOR calculation of the electric field distribution and partial experiments. The relationship between  $h/W$  and  $E/ER$  is calculated as a parameter of  $\epsilon$ . As a result, the FCME shows that the value of  $E/ER$  become higher as the value of the ratio  $h/W$  is increasing. High value of  $E/ER$  is also obtained by increasing the S/L, or lowering value of the  $\epsilon$ . We examined the domain inversion at the different value of  $E/ER$  with the same period ( $12 \mu\text{m}$ ) by using the high voltage pulser. As a result, domain inversion is well obtained depending on large  $E/ER$  value. So, it has become clear that the FCME is powerful technique with the narrower period, because of enhanced value of  $E/ER$ .

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