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Influence of light on the coercive field of repoled strontium barium niobate (SBN); the role of secondary repoling

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Abstract.

We have found that the application of light to strontium barium niobate (SBN) during electrical repoling stabilises the newly formed domains. This stabilisation becomes apparent when repoling the crystal back into its original domain direction as a change in the distribution of displacement current as a function of voltage. This appears to be the process underlying the other recent work in the area of optical control of domain structures for quasi phase-matching of nonlinear processes. We present an explanation for this effect in terms of the micro-domain structure of SBN. This model should aid in the search for new materials for optical periodic poling.

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The quasi phase-matching technique, achieved by periodically domain inverting certain ferroelectrics, is currently under intensive investigation by many research groups, as discussed in the recent review article by Houe and Townsend[2]. The results are very impressive: high efficiency second harmonic generation has been demonstrated, as well as optical parametric oscillation as far as 2.96 microns in the mid infra-red[3,4]. Several different techniques have been used to create periodically inverted materials including proton exchange, electrical field poling and, of relevance to this paper, all-optical and mixed optical-electrical methods. Kewitsch et al.[5] have demonstrated enhanced SHG using an all-optical method which exploits an internally generated space-charge field to create a head-to-head type domain structure in the material. Both Horowitz et al.[1] and Kahmann et al.[6] have used mixed electrical-optical methods to control domain structures, by using periodic light patterns and simultaneous application of an electric field along the c-axis, to control the repoling process. These two methods differ in the temporal sequence and magnitude of the applied fields: the method reported by Kahmann et al. has produced only 100 micron period structures, whereas Horowitz et al. have reported 1.5 micron size structures together with demonstration of enhanced SHG and parametric mixing. Although the conventional methods for producing QPM structures are very powerful, certain applications, such as backward wave optical parametric oscillation, would require sub-micron domain periodicities. Creating such fine structures by conventional techniques may prove extremely difficult, and so optical techniques may provide a valuable alternative.

In the following discussion we will have to refer to several successive stages of domain reversal by the application of electric fields. As there is no clear terminology to describe these stages we will use the following definitions. We shall restrict the use of the terms *poled* and *poling* to mean a process designed to place the crystal in a single domain **before** the start of an experiment, either by heating above the Curie temperature and cooling with an applied field, or by prolonged application of a field exceeding the coercive field. The term *repoling* will be used to describe the electrical reversal of the domain direction of a crystal **during** an experiment. The first and second *repolings* will be refered to as the *primary repoling* and *secondary repoling*, with associated *primary* and *secondary* coercive fields. Clearly the *secondary repoling* returns the polarization to its original *poled* direction.

In this letter we report a light induced change in the distribution of the *secondary repoling* displacement current after illumination of the crystal during the *primary repoling*. This method is essentially the same as that used by Horowitz et al.[1] to create QPM structures.
We present an explanation of the effect in terms of compensation of internal fields by photo-induced carriers.

Horowitz et al.[1] have reported several slight variants on their basic technique. In their latest work they used the following procedure. The crystal was poled into a single domain by using a large applied field; it was then illuminated with a 1mm wide strip of light normal to the c-axis in the centre of the crystal. The light pattern was formed by interfering two 10 mW beams from an argon-ion laser, operating at 514nm, focused through a cylindrical lens to produce an interference pattern with grating vector normal to the c-axis of the crystal. The primary repoling field was applied while the crystal was illuminated, and was larger than the primary coercive field of the crystal. The light was then blocked and a secondary repoling was performed, but, in this case the field used was smaller than the primary coercive field. The resulting structure was subsequently viewed using crossed polarizers.

In our work reported here, we have repeated this process using a SBN:61 crystal supplied by Deltronics Crystal Industries, but in our case we have also carefully monitored the displacement current during the repoling process. Horowitz et al.[1] did not report any measurements or analysis of the displacement current during the process. We initially poled the crystal into a single domain by heating it above the Curie temperature and cooling with an applied field, to reset the crystal to its original state and thus prevent deleterious effects due to electrical ageing[7]. By following the procedure in [1] we were able to obtain two seperate types of structures in our crystal. We believe that we have formed both a refractive index structure and a domain structure, produced by different mechanisms. We used the technique of two-beam coupling topography [8] to visualise the ferroelectric domain pattern directly, and used crossed polarizers or defocusing to see the refractive index structure. We used white light illumination to erase the refractive index structure to prevent distortion of the two-beam coupling image. Our investigations of this refractive index structure and its unusual properties will be published in a separate paper [9].

Our displacement current measurements have allowed us to understand more about the processes underlying the method of Horowitz et al.[1] (hereafter known as the Horowitz's method) for creating light controlled domain structures. We have recorded both primary and secondary repoling displacement currents with and without application of light during the primary repoling. Because the light controlled domain structures are only detectable after the secondary repoling we gain most information by analyzing the secondary repoling displacement current as a function of illumination during the primary repoling. During the primary repoling the electric field was ramped from 0 to 4kV/cm in 30 seconds. Illumination, when used, was provided by an argon ion laser utilising the 514nm line. Secondary repoling current curves were obtained by ramping the field from 0 to 4kV/cm in 225 seconds, and recording the displacement current with a sensitive current to voltage pre-amplifier. Typical secondary repoling current curves are shown in Fig. 1; curve A was taken without illumination during the primary repoling, whilst curve B was taken with an optical field of 60mW/cm^2 irradiance.

Comparison of the curves shows that the displacement current flowing at low fields is very much larger if no light has been applied during the primary repoling. This result holds the key to understanding Horowitz's method. During the primary repoling the whole crystal is repoled into a single domain, then during the secondary repoling the applied field is chosen to be slightly smaller (in this case about 400V/cm less would be suitable) than the secondary coercive field. This means that in the dark regions of the crystal, some domains will flip back to their original direction, whereas in the illuminated regions very few domains will flip back. Thus we can see that illumination during the first stage stabilises the newly formed domains, allowing us to create a spatial domain structure. By measuring the total charge flowing up to the maximum secondary field value we obtain the switched polarization volume, in this case about 15.5% (assuming a maximum secondary field strength of 1.7kV/cm) without illumination during the primary repoling, compared with a switched volume of 3.5% after primary illumination. So the structure is poled / partially depoled rather than being properly poled / reversed poled. However, it ought to be possible to optimize the poling contrast by adjusting the experimental parameters. It is found that the primary coercive field is larger than the secondary coercive field, with values of 2.8kV/cm and 2.2kV/cm respectively. Such behaviour is common in many ferroelectrics, [10], and manifests itself as a biased hysteresis loop.

We now present a tentative explanation for this result. It is well known that SBN contains a rich structure of micro-domains[11]. The microscopic variation in the domain structure implies that charges must exist in the bulk of the material to compensate the variation of the polarization. These charges (shown schematically in Fig. 2) stabilise the domain configuration of the crystal, and will oppose any repoling. We consider the illuminated and unilluminated parts of the crystal separately in what follows.

In the unilluminated regions of the crystal the primary repoling will cause micro-domains to flip through 180 degrees because the applied field is considerably larger than the primary coercive field. The bulk charges (which are unable to move) now act to destabilise the new domain structure, so that during the secondary repoling the domains will flip back to the original direction at fields very much lower than the secondary coercive field.

The situation in the illuminated regions is rather different because during the primary repoling photo-carriers will be excited which can neutralise the effects of the bulk charges, so that the domains will not flip back so easily at low voltages. Thus we are able to explain the observed current curves, and if the maximum secondary field is chosen carefully, we can arrange for very few of the illuminated micro-domains to flip back whilst many unilluminated micro-domains will.

Recent work by Chao et al. [12] has also shown that light can influence the coercive field after electrical domain inversion in $LiTaO_3$. They followed a very similar process except that light was applied during the secondary repoling. They found that the secondary coercive field was smaller than the primary one and that after some time it recovered towards the primary value. Furthermore, they found that this recovery could be hastened by applying light to the sample. This similarity also highlights the role that light controlled secondary repoling effects might have in creating periodically domain inverted material by optical routes. It also suggests that $LiTaO_3$ is a strong candidate for optical poling.

In conclusion, we have investigated the secondary repoling characteristics of SBN with and without light during the primary repoling. This process corresponds to Horowitz's method for producing domain structures. We have made displacement current measurements that allow us to identify the mechanism as being a light induced stablisation process, and have presented an explanation based on the micro-domain structure of SBN. This effect appears to be closely allied to that seen by Chao et al. in $LiTaO_3$. We believe that such light induced secondary repoling effects may be very important in the further development of optical periodic poling, and in extending QPM to ultra-small periods. Additionally, the understanding we formed about the process in SBN should allow us to identify other materials, possibly including $LiTaO_3$, which may prove suitable for optical poling.

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Figure 1

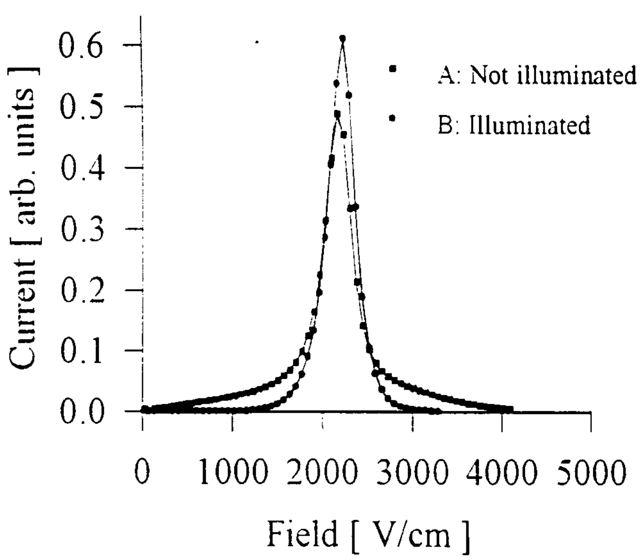
Caption:

Secondary repoling current curves. Note that the current at low fields is smaller when the crystal has been illuminated during the primary repoling.

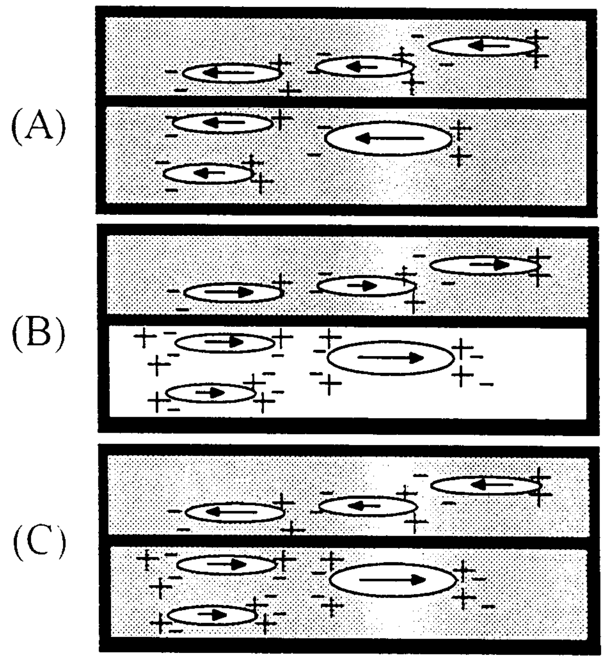
Figure 2

Caption:

In the initial poled state (A) compensating charges in the bulk of the crystal neutralise the variation of polarization due to micro-domains. During the primary repoling (B) all of the domains flip, but photo-carriers in the illuminated regions neutralise the destabilising effect of the original compensating charge. During the secondary repoling (C) the combination of the destabilising fields and the externally applied field causes some of the domain to flip. Whereas in the previously illuminated regions the destabilising field is smaller and so domains do not flip back so easily at the fields used.



0.6
0.5
0.4
0.3
0.2
0.1
0.0
Current [arb. units]
A: Not illuminated
B: Illuminated
0
1000
2000
3000
4000
5000
Field [V/cm]


(A)
(B)
(C)