The effect of the ferroelectric domain walls in the scanning near field optical microscopy response of periodically poled $Ba_2NaNb_5O_{15}$ and $LiNbO_3$ crystals

2009 J. Phys.: Condens. Matter 21 042201

(http://iopscience.iop.org/0953-8984/21/4/042201)

IOP FTC ▶▶▶

IOP PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **21** (2009) 042201 (4pp) doi:10.1088/0953-8984/21/4/042201

FAST TRACK COMMUNICATION

The effect of the ferroelectric domain walls in the scanning near field optical microscopy response of periodically poled $Ba_2NaNb_5O_{15}$ and $LiNbO_3$ crystals

T P J Han[1], F Jaque[1], J Lamela[2], D Jaque[2], G Lifante[2], F Cusso[2] and A A Kamiskii[3]

[1] Department of Physics, University of Strathclyde, Glasgow G4 0NG, UK
[2] Departamento de Física de Materiales, Universidad Autónoma de Madrid, 28048-Madrid, Spain
[3] Institute of Crystallography, Russian Academy of Sciences, Moscow 119333, Russia

E-mail: t.han@strath.ac.uk

Received 4 August 2008, in final form 21 November 2008
Published 15 December 2008
Online at stacks.iop.org/JPhysCM/21/042201

Abstract
A study of $Ba_2NaNb_5O_{15}$ and $LiNbO_3$ crystals with periodic ferroelectric domain structures using the scanning near field optical microscopy technique is reported. Optical contrast is observed in the regions of ferroelectric domain boundaries and it is analysed using beam propagation method modelling. This reveals that the optical contrast, a consequence of changes in the refractive index, is not due to variation of the waveguide-coupling efficiency, and supports the hypothesis that it is associated with the domain array, which is related to the size of the domain.

(Some figures in this article are in colour only in the electronic version)

Since the discovery of ferroelectricity by Valasek in 1921 [1] the characteristics and the nature of the ferroelectric domains have received a great deal of attention because of their relevance in practical applications. The presence of domain boundaries could limit the potential optical applications of ferroelectric systems as they can act as scattering centres. However, detail understanding of the formation mechanisms of these domain boundaries and their local environment could give further insight into the physics governing ferroelectricity and local structural characteristics. Although formation and dynamics of ferroelectric domains have been extensively studied in different ferroelectric materials, there is still a long way from a full understanding of the properties of the domain boundaries. $LiNbO_3$ and $Ba_2NaNb_5O_{15}$ are ferroelectric crystals extensively used in photonics devices because of their high nonlinear and electro-optic coefficients. In recent years, the viability of creating periodic ferroelectric domain structures using different processes has extended the applications of these materials in the photonic field even further [2, 3]. In a recent study on $LiNbO_3$ crystals of congruent composition, using the scanning near field optical microscopy (SNOM) technique Kim *et al* [4] observed unexpected optical contrast in the regions of a single ferroelectric domain wall created by room temperature electric field pooling. As the refractive index has centrosymmetric property it is not expected to present any difference between the two sides of the domain boundary. So, the high optical contrast found was suggested to be related to the presence of local strains and electric fields. In particular, it was assumed that the local strain was associated to the intrinsic defect complexes such as lithium vacancies and the so-called antisites where the niobium ions are located in lithium vacancies. The structure of these complexes was considered to be different at either side of the domain boundary. This asymmetry is, therefore, assumed the cause of the local lattice strain. The magnitude of this local perturbation is maximum closest to the boundary and decrease over several microns away from the domain wall. This local strain is responsible of the change in the local refractive index, Δn. Under this assumption, the ferroelectric domains operate as a light pipe producing a large optical contrast. By means of beam propagation method (BPM) and finite difference time domain (FDTD) method Kim *et al* [4] obtained a value of the order of 10^{-3} for Δn over a distance of ~20 μm from the domain boundary [4]. In another study of periodically poled $Ba_2NaNb_5O_{15}$ crystals a modulated scanning near field optical microscope (SNOM) optical contrast has also been reported with the same periodicity as the domain structure [5]. The authors analysed their observation using BPM method and suggested that the ferroelectric periodic domains operates as an array of planar waveguides with a modulation of the refractive index of $\Delta n \sim 10^{-4}$. Although, this refractive index modulation is expected to be strongly dependent of the domain size (periodicity of the ferroelectric structure), this fact has not been yet investigated. It is evident that a detail study of the boundary-induced refractive index singularities dependence on the ferroelectric domain size can provide useful information on the boundary region of ferroelectric structures.

The aim of this letter is to report on a quantitative SNOM study in $Ba_2NaNb_5O_{15}$ (BNN) and $LiNbO_3$ (LNB) crystals, with periodic ferroelectrics domains structures created during the crystal growth procedure, as a function of the ferroelectric domain size.

It is generally accepted that periodic poled structures are easier to form when impurities are added in the melt of the Czochralski growth technique. In this work BNN crystals doped with 0.5 mol% of Nd^{3+} ions (BNN:Nd) and LNB crystals of congruent composition codoped with equal concentration of 0.5 mol% of Er^{3+} and Yb^{3+} ions (LBN:Er:Yb) were grown along the a-axis in air by the 'off-centred' Czochralski method. Samples of 1 mm thickness were cut and polished to optical grade for the SNOM measurements. The presence of ferroelectric domains in the samples is verified by means of selective chemical etching treatments.

SNOM images were obtained with a Nanonics Imaging Ltd model MultiView 200™ working in non-contact tapping mode. A continuous-wave laser beam from a 532 nm doubled Nd:YAG laser was coupled into the SNOM probe and data were collected by working in either the reflection or transmission mode. Under these configurations the sample surface is illuminated through the fibre probe with a 100 nm tip size and the reflected or transmitted light is collected through a 10× long work distance objective and focused onto an avalanche photodiode (APD). Figure 1(a) displays the 3D SNOM transmission image of the BNN:Nd sample showing a clear optical intensity modulation ΔT. A fast Fourier transform (FFT) analyses confirmed the observed periodicity of 4.8 μm. The calculated optical contrast $\Delta T/T$, where T is the transmission intensity obtained from the transmission intensity profile, is around 20%. It has been verified that the magnitude of optical contrast, $\Delta T/T$, remains constant for sample thickness between 1 and 3 mm.

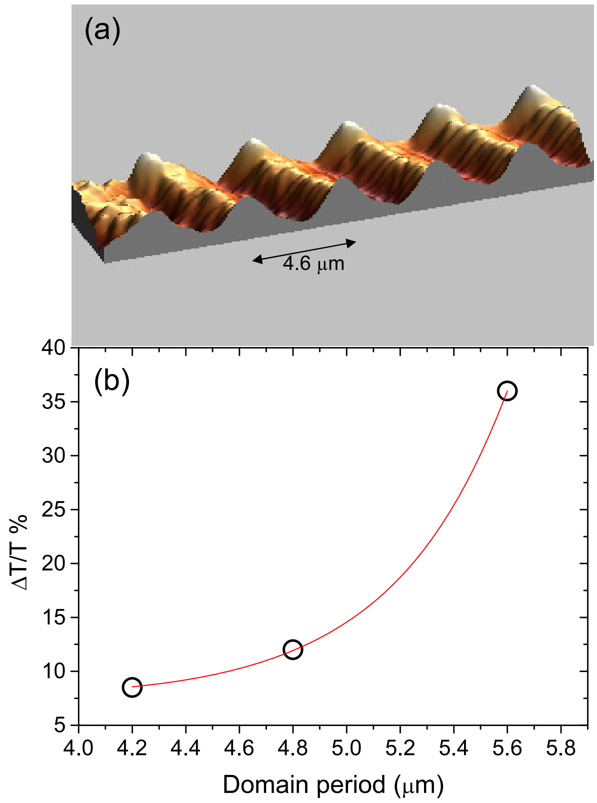

Figure 1. (a) Transmission 3D SNOM image of doped $Ba_2NaNb_5O_{15}$:Nd sample. (b) Variation of the transmission optical contrast with the periodic domain value at various positions on the sample surface.

The ferroelectric domain period was observed to vary between 4 and 6 μm. This lack of periodicity is an undesirable feature in practical applications but it can be useful to study the dependence of the refractive index modulation with the size of the ferroelectric domains. Figure 1(b) shows the transmission optical contrast ($\Delta T/T$) dependence with the domain size (domain period). As can be seen a significant variation of $\Delta T/T$ from 8% (domain period 4 μm) to 35% (domain period 5.6 μm) is observed. The topography of the sample surface obtained simultaneously with the SNOM image shows a roughness <5 nm without any defined structure indicating that the changes observed in the intensity transmission signal cannot be attributed to any topographic effects. Using the SNOM in reflection mode a similar, but much weaker optical contrast $\Delta R/R$ modulation was also observed. This small optical contrast has been attributed to waveguiding effect: the ferroelectric domains are acting like waveguides so that the reflected light from the back surface of the sample is guided back to the front surface increasing the reflected light intensity of front surface [6].

The SNOM measurements of the LNB:Er:Yb sample were obtained in the reflective mode as it produced a much superior contrast than the transmission mode. This could be due to the fact that $LiNbO_3$ is very sensitive to its growth condition and the off-centred growth technique might produce non-parallel domains through the whole boule resulting in poor quality transmitted light. The backscattering reflected

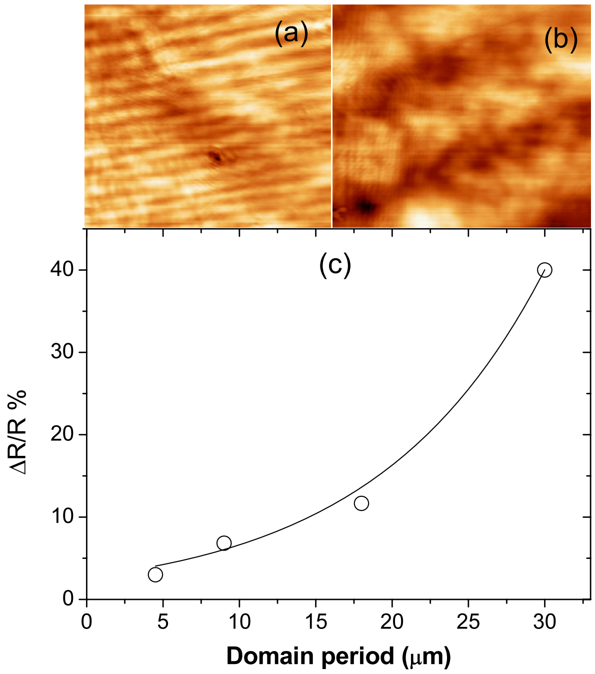

Figure 2. Reflected 2D SNOM images for two different domain periods (a) ~5 μm and (b) ~18 μm obtained in doped $LiNbO_3$ crystals. (c) Variation of the reflected optical contrast with the periodic domain value at various positions on the sample surface.

intensity, R, showed a high optical contrast modulation, ΔR, with the same period as the ferroelectric domain revealed by etching. However, at variance with the BNN:Nd crystal, a large variation of the domain period was available in this sample: from a few microns up to 30 μm. Figures 2(a) and (b) show the reflected 2D SNOM image for areas of the sample where a greater percentage of domain periodicity with 4.8 μm and ~18 μm were measured. The optical contrast, $\Delta R/R$, obtained is ~3% and ~10% respectively. In figure 2(c) the variation of $\Delta R/R$ as a function of the domain period is presented. A nonlinear variation of $\Delta R/R$ from 3% to 40% is recorded in the periodic domain range from 5 to 30 μm. The results obtained for the two crystals, considering an equal period variation range, seem to indicate the periodic effect is greater in the BNN:Nd sample. The origin of this is not known and is currently under further studies.

The optical contrast observed in periodic poled BNN:Nd crystals is analysed by considering the ferroelectric domains as an array of planar waveguides structures [4, 5]. Figure 3 shows calculation based on the BPM model [5] using a refractive index modulation of $\Delta n \sim 10^{-4}$ and a domain period close to 5 μm. The result shows that for domain periods range between 4 and 8 μm the variation in $\Delta T/T$ is only <3% which is much lower than the ~30% observed experimentally. This demonstrates the observed optical contrast change with periodicity is not simply due to the changes in size of the waveguide, i.e. domain. If one considered the proposition that the domain boundary produced strain and/or deformation, hence the changes in refractive index associated to domain boundaries, Δn, would extend tens of microns from the boundary wall [4]. This implies that, in the case of a

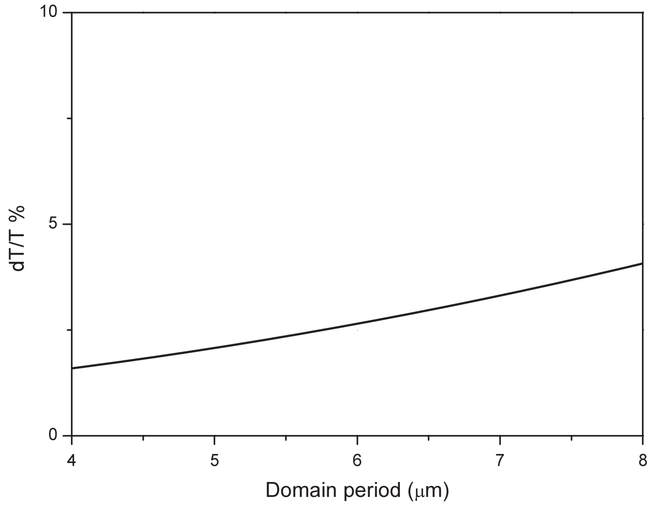

Figure 3. BPM simulation of the dependence of the transmission optical contrast in a waveguide array with the waveguide size.

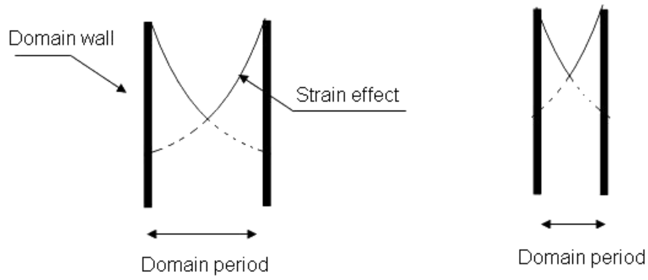

Figure 4. Diagram showing the variation of the refractive index between two boundary walls of two different domain periods.

micrometric array of such domain boundaries, Δn variation between two domains walls should be a function of the wall–wall distance. This supposition is illustrated in figure 4, showing that the Δn values becoming weaker for smaller distances between the boundary walls. A more refined and quantitative study of the variation of Δn with distance between domains walls would need more experimental information, expanding the experimental range of the domain period studied here. However the data obtained in this study indicate clearly that the effect of the ferroelectric walls extends at least up to 30 μm in $LiNbO_3$.

In conclusion, the SNOM optical contrast data obtained in doped BNN and $LiNbO_3$ crystals with periodic domain ferroelectric structures shows a dependency with the domain period. A BPM model reveals that this behaviour is not due to a variation of the waveguide-coupling efficiency related to the size of the waveguide. Whereas, the observed data seems to support the hypothesis that the domain boundaries induced a perturbation close to the boundaries which can extends to tens of microns from the wall and hence the magnitude the refractive index modulation associated to the domain array is related to the size of the domain.

This work has been supported by the Spanish Ministerio de Investigación e Innovación (proyect MAT2005-05950

MOISES) and by the Comunidad de Madrid (proyect S-0505-TIC-0191 MICROSEREES) and Universidad Autónoma de Madrid.

References

[1] Valasek J 1921 *Phys. Rev.* **17** 475
[2] Moue M and Townsend P D 1995 *J. Phys. D: Appl. Phys.* **28** 1474
[3] Pruneri V, Butterworth S and Hann D C 1996 *Appl. Phys. Lett.* **69** 1029
[4] Kim S and Gopalan V 2005 *Mater. Sci. Eng.* B 12091
[5] Lamela J, Ródenas A, Lifante G, Jaque D, Jaque F and Kaminskii A A 2008 *Laser Phys. Lett.* **5** 291
[6] Radko I P, Volkov V S, Bozhevolnyi S I, Henningsen J and Pedersen J 2005 *Laser Phys. Lett.* **2** 440