

Image and hologram fixing method with $\text{Sr}_x\text{Ba}_{1-x}\text{Nb}_2\text{O}_6$ crystals

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We demonstrate a new fixing method with $\text{Sr}_x\text{Ba}_{1-x}\text{Nb}_2\text{O}_6$ crystals. This is done by inducing ferroelectric domains in the crystal in real time, by a screening mechanism. Images or their holograms can be recorded in small separate locations in the crystal. This crystal pixelization permits the recording of many pictures and their individual readout and erasure.

New methods for fixing of images and holograms in crystals can provide an important basis for new optical image processing and memory applications. The currently known fixing methods¹⁻⁵ are limited, and their repeatability and effectiveness for long times are not clear. They are based on ideas from approximately twenty years ago¹⁻³ of using photorefractive crystals and forming in them ionic changes by light-induced space-charge fields. This can be accomplished,¹ for example, by applying an electric field in the direction opposite to the crystal polarization or by heating the crystal. Then the local field (the sum of the external and space-charge fields) causes a partial switching of the polarization. The obtained spontaneous polarization of the domain grating must be oriented longitudinally, i.e., along the space-charge field and the grating wave vector. These methods have an inherent limitation owing to the built-in depolarization electrostatic energy in the domain walls, since $\nabla \cdot \mathbf{P} \neq 0$ (\mathbf{P} is the spontaneous polarization) between successive antiparallel domains. This limits the long-term stability of the domain structures, especially for short grating wavelengths.

In this Letter we report on a new reversible fixing method with $\text{Sr}_{0.75}\text{Ba}_{0.25}\text{Nb}_2\text{O}_6$ crystals. Our fixing procedure uses an applied electric field on the crystal and a simultaneous illumination of a small part of it with a light pattern. We have demonstrated direct image and hologram recording. The method is different from previous processes in the following points: (1) Screening is used for affecting or canceling an applied field for domain inversion, with the possibility of transverse domain gratings (with respect to the wave vector). In this case the domain wall depolarization energy is eliminated ($\nabla \cdot \mathbf{P} = 0$). (2) The induced domain gratings can have an unusual wave vector (modulation) direction, perpendicular to the c axis, with periodicities as short as the order of $1 \mu\text{m}$. (3) There is the possibility of obtaining fixing in crystals that are not photorefractive. (4) There is the possibility of storing many different holograms in small separate regions in the crystal. Each hologram can be separately erased or stored in a few seconds.

Recently,^{6,7} with our fixing method, we produced periodic domain structures (domain gratings) with alternating orientations in $\text{Sr}_x\text{Ba}_{1-x}\text{Nb}_2\text{O}_6$ crystals for quasi-phase matching in second-harmonic generation. We were able to obtain controllable narrow-band and broadband second-harmonic generation. The modulation direction perpendicular to the c axis was essential in this experiment.

The experimental setup for the fixing process is shown in Fig. 1. For the writing light beams we used an argon-ion laser with a wavelength of 514.5 nm, an extraordinary polarization, and powers of 5 to 30 mW. The crystals were poled $\text{Sr}_{0.75}\text{Ba}_{0.25}\text{Nb}_2\text{O}_6$. One of them was undoped without pronounced photorefractivity, with dimensions of approximately $5 \text{ mm} \times 5 \text{ mm} \times 5 \text{ mm}$. Another one, nominally doped with 0.05 wt. % Ce, was photorefractive, with dimensions of $6 \text{ mm} \times 6 \text{ mm} \times 1 \text{ mm}$ and a c axis along one of the 6-mm sides. The experimental data given here were obtained with the second crystal. In the direct image recording, we used only one beam of the argon-ion laser, and an image was projected onto a small area of the crystal by a lens. The fixing caused permanent phase patterns in the crystal. In the hologram fixing experiment, another reference beam was used to form in a small area of the crystal an interference with the image carrying beam. The crystal was located close to the Fourier plane of the image. In both cases

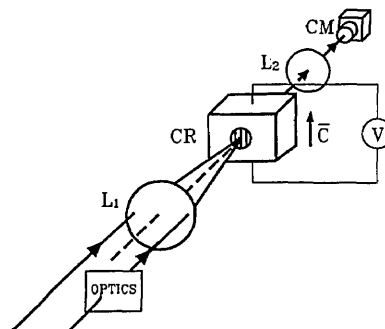


Fig. 1. Experimental setup for the fixing process. CR, crystal with the indication of the c axis; L_1 , lens with a focal length of 10 cm; L_2 , lens; CM, camera.

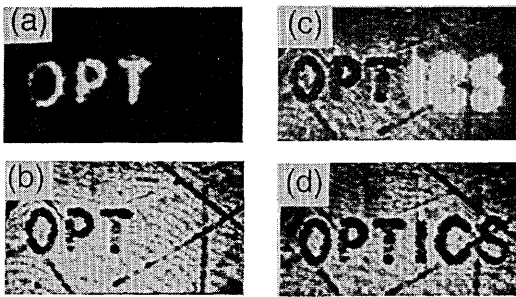


Fig. 2. Fixing of direct images in the crystal: (a) the original first image (after passing the crystal), (b) the reconstruction of the recorded image, (c) a second image (added to the first), (d) the reconstruction of the two images that were stored on the crystal.

(direct image and hologram recordings) the fixing was carried out in two stages: First, an external voltage was applied along the c axis with an opposite polarity to that used to pole the crystal. The voltage was gradually increased from 500 V to 2 kV in 10–15 s until a phase distortion was seen on the image beam after it passed the crystal. The needed strength and duration of the applied voltage were shorter as the intensity of the input light was higher. Then the writing beams were blocked and the polarity of the applied voltage was inverted. The voltage on the crystal was raised from 0 to 800 V in 5 s and then disconnected. During the reconstruction of the information a low voltage (≈ 100 V) was applied, to increase the efficiency. This voltage, needed to form a refractive-index difference between the antiparallel domains, caused an immediate growth in the intensity of the reconstructed image as a result of the electro-optic effect. This voltage was not always needed in the doped crystals.⁶ The fixing was stable, and no significant change in the visibility of the reconstructed image was seen after we illuminated the crystal for 1 h with an argon-ion laser beam with an intensity of 20 mW/mm^2 , turning it off for 12 h and then turning it on for another hour. The fixing in a specific volume in the crystal could be erased by illumination of the crystal with a 20-mW argon-ion laser beam while an external voltage of 2 kV was applied for approximately 15 s.

In the first part of the experiments we stored an image in the crystal. After the fixing, the beam that contained the image was blocked, and the fixed image was observed by a Gaussian probe laser beam (He–Ne). The fixing process created changes in the crystal that resulted in spatial modulation of the phase of the probe beam. In order to observe these changes, we placed the CCD camera slightly out of the image plane of the crystal, so that the phase structure was converted into an intensity pattern. In the same manner we could see the phase pattern by illuminating the crystal with a white light instead of the laser beam. Figure 2 shows the original image and the reconstructed image. The size of the image diameter in the crystal surface was $\sim 100 \mu\text{m}$; however, larger or smaller image sizes could be stored. We have noticed that the fixing process caused significant changes in the crystal structure only in the regions that were illuminated

during the time that the external field was applied. We used this property to store another image (after the first image was stored) in a different location of the crystal [Fig. 2(c)]. We added the new image to the crystal by projecting it on the desired location and performing the fixing process as described above. No significant degradation of the reconstruction quality of the first image was observed after the fixing of the second image [Fig. 2(d)].

In the hologram fixing experiments, we recorded the interference of the image-carrying beam with a reference beam that had a diameter of 1 mm. The wave vector of the fringe pattern was approximately normal to the c axis (unlike for Ref. 1). Then the space-charge field built by the photorefractive process was approximately perpendicular to the applied field. The period of the grating was $\sim 2 \mu\text{m}$; however, holograms with smaller periods could be fixed. Before the fixing step, we observed a very weak diffraction of the reference beam in the direction of the signal beam, when the input beams had extraordinary polarizations. We think that this coupling is caused by the photogalvanic effect,^{8,9} which explains why no significant diffraction could be seen when the input beams had ordinary polarizations. After the fixing process was performed, the signal beam was blocked, and the reconstructed image could be seen. The original and the reconstructed images are shown in Fig. 3. The diffraction efficiency in this case was $\sim 1\%$ (for a crystal width of 1 mm) and slightly increased as we increased the grating's wavelength.

We have also stored two holograms in different locations of the crystal. However, when the fringe gratings had small periods ($\approx 2 \mu\text{m}$), we observed a significant decrease of the reconstruction efficiency of the hologram that was first stored, unless we used strong light powers for the fixing ($\approx 150 \text{ mW}$). In this case, the voltage needed for the first stage of the fixing process was less than 1 kV. High applied voltages on the crystal ($> 1.5 \text{ kV}$) caused the erasure of holograms with small

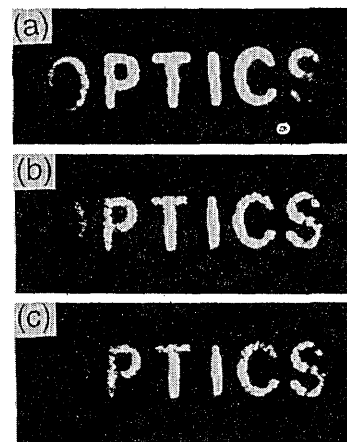


Fig. 3. Fixing of the hologram in the crystal: (a) the original image, (b) the reconstructed image, (c) the reconstructed image after the crystal was illuminated with an argon-ion laser for 2 h. The intensity of the incident light was 20 mW/mm^2 .

grating wavelengths even without illumination. We also stored several holograms in different locations of the crystal by repeating stage 1 of the fixing process for each hologram separately, without changing the polarity of the applied voltage. Then we performed the second stage of the process, which simultaneously fixed all the holograms without any mutual damaging effect. This fixing could be obtained with low light power. We could also store holograms with grating wave vectors that were approximately parallel to the c axis. In this case high diffraction efficiencies of 2% to 6% were observed for gratings with long periods ($>10 \mu\text{m}$). However, gratings with periods of $\sim 2 \mu\text{m}$ resulted in low diffraction efficiencies ($\approx 0.15\%$). The diffraction efficiency could be enhanced temporarily by application of a strong voltage to the crystal; however, this voltage caused the erasure of the stored hologram.

We believe that the fixing is based on screening. The applied field is screened in the regions with strong light intensities by the charge separation enabled by the photoionized charges. The screening produces strong internal fields, causing nonuniform distribution of the applied voltage. These fields, which follow the light intensity pattern, result in a nonuniform inversion of the spontaneous polarization. Even after the applied voltage is turned off, the strong internal field may cause further domain inversions. The general understanding of the nature of domain inversion in the presence of internal fields and photoinduced charges is an interesting topic for further research. The role of free carriers on the domain structure in ferroelectric crystal was raised a long time ago.⁸

The indications that the fixing mechanism is based on screening are the following: (1) We were able to obtain good fixing in crystals without a pronounced photorefractivity. Moreover, the fixing was obtained for direct images that were projected on the crystal, where only one beam illuminated the crystal and the light intensity gradients were low. Then the standard photorefractive effect is very weak. (2) We have stored holograms with wave vectors that are approximately perpendicular to the applied fields. In these cases the standard space-charge photorefractive field is normal to the applied field so that it cannot trigger the domain inversions. (3) When the light illuminated a layer in the crystal that was parallel to the applied field there was no observed change in the crystal after the fixing process. The screening effect is simply not effective in this case. However, when the light layer was normal to the applied field, we obtained a strong fixing with phase distortions and light scattering in the layer.⁶ In fact, weak fixing could be achieved in this case even without the use of external fields if the power of the incident light was above 100 mW (the width of the layer was ~ 0.2 mm). This is due to heating of the crystal by the light to the phase transition temperature (45°C in $\text{Sr}_{0.75}\text{Ba}_{0.25}\text{Nb}_2\text{O}_6$ crystals¹⁰), where the coercive field is negligible and the internal photogalvanic field^{8,9} becomes sufficient for inducing domain inversions. One can use this property to obtain fixing without

applying external fields. We believe that such fixing was reported in Ref. 3. However, the fixing obtained by this method is weak and has a limited lifetime.

There are two other factors in the fixing process: (1) The photogalvanic effect,^{8,9} which, in the case when the grating wave vectors are approximately perpendicular to the applied field, causes internal fields in the direction of the applied field. These fields might have been a cause for domain switching. However, when we used perpendicular polarizations for the two writing beams, no diffraction of the reference beam toward the image beam was observed before and after the fixing. Such a diffraction is expected for crystals with a large photogalvanic effect.⁹ (2) Photoinduced free carriers compensate for the depolarization fields by conduction⁸; thus we get different behavior of the domain inversions in the dark and in the illuminated regions of the crystal.

Our understanding that the fixing process involves domain inversion is based on the following two reasons: (1) Fixed gratings in our method were used to provide quasi-phase matching for efficient second-harmonic generation.^{6,7} This necessitated a domain inversion that provided a domain grating with a modulation direction perpendicular to the c axis. We were able to obtain controllable narrow-band and broadband harmonic generation in a prespecified wavelength region. We could change this wavelength in a short time by performing a new fixing with a proper period. (2) No significant fanning (a strong asymmetrical light scattering in photorefractive crystals) was observed after the fixing was performed. This was a drastic change with respect to the prefixing stage, in which the fanning was very strong.

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