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APERIODIC OSCILLATIONS IN THERMAL FOCUSING BY SINGLE CRYSTALS OF LEAD MAGNESIUM NIOBATE

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Abstract Optical bistability in monocrystalline lead magnesium niobate, $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$, (PMN) has been studied at temperatures near the ferroelectric phase transition under 514.5 nm illumination. At moderate laser power densities above a threshold but less than 100 kW/cm^2 , this material exhibits reproducible aperiodic oscillations following the rapid switching on of a constant intensity Gaussian laser beam. With increasing incident power the oscillations change from the symmetric type observed in lanthanum modified lead zirconium titanate (PLZT) to a sequence of rapid jumps followed by a slow relaxation between two metastable thermal lens configurations, similar to those observed in ceramic samples of PMN at low power densities (about 1 kW/cm^2). The duration and number of the oscillations depend reproducibly on incident power and ambient temperature, with different spatial structures from those observed in ceramic samples, depending not only on incident beam power but also on incident beam width and wavefront curvature.

INTRODUCTION

Recently transverse optical bistability has been studied in ferroelectrics near their Curie temperatures, where the thermo-optic coefficient $(1/n)(dn/dT)$ (where n = refractive index and T = temperature) is large (e.g. ca. 9×10^{-5} for PMN)¹. In particular, a ceramic wafer of titanium-doped lead magnesium niobate, $\text{Pb}(\text{Mg}_{0.3}\text{Nb}_{0.6}\text{Ti}_{0.9})\text{O}_3$, illuminated by a laser beam which is rapidly switched to a steady power level, produces a complex dynamic response², involving a sequence of sudden shifts in the radial intensity distribution within the transmitted and reflected beams, which coincide with rapid switching of the total irradiances of the transmitted and reflected beams, as well as in the on-axis transmitted irradiance. These rapid changes occur at geometrically increasing times, separated by intervals during which slow relaxation of both transverse patterns and irradiances occur.

In this paper we report on the static and dynamic response of monocrystalline undoped PMN. This material differs significantly from the ceramic doped samples previously studied. The effective optical absorption is significantly lower than in the ceramic, since it is not enhanced by scattering at domain boundaries and voids, and the thermal conductivity is much larger. The optical power densities required to obtain optical bistability are therefore between one and two orders of magnitude higher than in the previous work ($10 - 100 \text{ kW/cm}^2$ as against $\approx 1 \text{ kW/cm}^2$), necessitating a more tightly focussed incident beam from a c.w. laser. The samples used were thicker ($0.68 - 1.40 \text{ mm}$) than ceramic sample (0.43 mm). The static and dynamic responses were qualitatively different in different regimes defined by the sample thickness and the beamwidth and wavefront curvature of the incident light.

RESULTS

Experimental

Single crystals of PMN with large parallel facets parallel to the $(100)_{\text{cub}}$ and $(110)_{\text{cub}}$ planes and polished to optical quality and thicknesses between 0.68 and 1.40 mm were illuminated at a wavelength of 514.5 nm . The sign of the thermo-optic coefficient responsible for thermal lensing in PMN was determined by using a collimated incident beam of diameter *ca.* 0.7 mm . Above a threshold laser power of approx. 200 mW this beam was focussed beyond the sample, indicating that the thermo-optic coefficient is positive, in agreement with measurements reported at a wavelength of $632.8 \text{ }\mu\text{m}^1$. As the laser power was slowly increased, the far field transmitted beam profile changed from the approximately Gaussian intensity profile of the incident beam to a thermal lens pattern with a bright outer ring and a sequence of bright and dark regions at the centre. When the incident beam was switched to a steady power level, the thermal lens pattern reached steady state within 1 second.

Figure 1 shows aperiodic asymmetric relaxation pulsations of the on axis far field transmitted intensity produced with a focussed incident beam (incident spot diameter: $30\text{--}50 \text{ }\mu\text{m}$) above threshold power, having a time dependence similar to that observed in ceramic PMN. Below the threshold for thermal focussing, the on-axis transmitted intensity still showed aperiodic oscillations but of lower relative amplitude and approximately symmetric shape similar to that observed recently in PLZT³. The threshold power (W) for thermal lensing was within a factor of 2 of that for the collimated beam for constant sample thickness, although the incident power density (W/cm^2) at the sample was more than two orders of magnitude greater. The power threshold for thermal focussing (somewhat higher than that for oscillation) varied inversely with the crystal thickness: e.g. 100 mW for 1.40 mm , 300 mW for 0.74 mm and $>300 \text{ mW}$ for 0.68 mm , with an incident spot diameter of $40 \text{ }\mu\text{m}$. These two facts suggest that total absorbed laser power may be a threshold parameter rather than incident power density.

When the sample was placed at or slightly in front of the beam focus, the static and dynamic responses of both the transmitted and reflected beams were qualitatively similar to those previously observed in samples of ceramic PMN². During slow

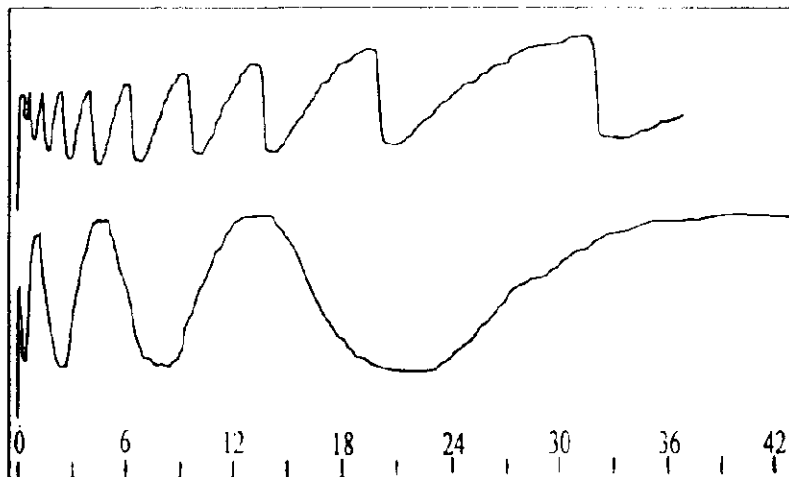


FIGURE 1: Relative far field central intensity transmitted by a PMN crystal 1.40 mm thick vs. time (s). Steady incident power: 90 mW (lower curve) and 230 mW (upper curve).

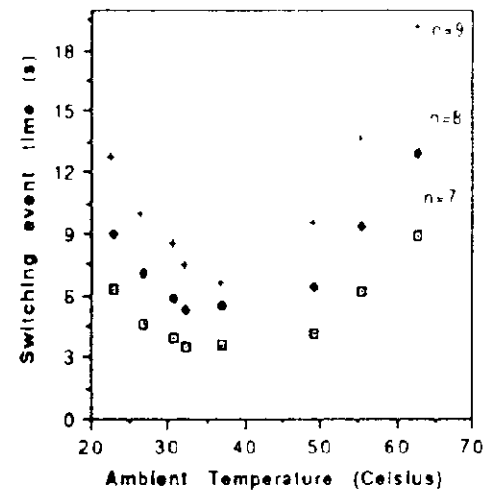


FIGURE 2: Switching event time t_n ($n = 7 - 9$) vs. temperature for 1.40mm PMN crystal.

upward ramping of the laser power to a given level, a number of jumps occurred in the patterns of both transmitted and reflected beams which was equal to the number of jumps which occurred after sudden switch-on to the same power level. The same number of jumps occurred when the power was ramped down again but in the opposite sense and at correspondingly lower power levels (i.e. the patterns showed multistable hysteresis loops).

With the sample >2 mm behind the focus, the transmitted beam was focussed to a small bright spot surrounded by several closely spaced bright and dark fringes. The reflected beam produced a pattern of broad circular fringes which contracted towards the centre of the pattern and disappeared successively, both during slow upwards ramping of the power and following rapid switch-on. These fringe contractions occurred in rapid jumps at increasing time intervals when the sample was near the beam waist but became more continuous as distance from the beam waist increased. With the sample about 2 mm behind the focus, the response patterns varied qualitatively with incident laser power. When power was slowly ramped upwards, the pattern switched from the concentrated post-focus pattern to the expanded pre-focus pattern, as the original beam focus was pulled into the sample by thermal focussing.

As shown in Figure 2 (for steady power = 230 mW), the number and switching time of the aperiodic oscillations varied systematically with ambient temperature. From 20°C to ca. 40°C, switching times decreased and the number of oscillations increased. The temperature dependence was reversed from 40°C to 70°C.

Spatial Ring Soliton Interpretation

The characteristic shape of the thermal lens pattern, consisting of a bright ring surrounding a lower intensity broad central spot, can be explained qualitatively by the folding of an outer annular region of the incident beam over the inner section due to the fact that deviation due to the thermal focussing effect is greatest for

incident rays with intermediate convergence angles. The oscillating thermal lens pattern produced by a defocussing thermochromic liquid contains a broad dark ring⁴ whereas some PMN patterns show a very narrow dark ring. Quantitatively the patterns result from the solution of the slowly varying amplitude equations for the forward and backward travelling waves in a Fabry-Perot cavity, coupled via the thermo-optic coefficient and the absorptive heat source to the heat equation subject to convective boundary conditions⁵. This system of equations is similar to the perturbed cylindrical Nonlinear Schroedinger Equation. It has been shown that this equation has solutions in the form of ring-shaped spatial solitons, which are bright for a focussing medium and dark for a defocussing medium and in both cases are stable under several different types of perturbations⁶. The temporal oscillations may therefore be interpreted as cycles of formation, growth and breakdown of spatial ring solitons, as the rising temperature tunes the Fabry-Perot cavity through a number of resonances. A one-dimensional model of this process yields a finite sequence of oscillations for both positive or negative thermo-optic coefficients⁷ but a finite transverse dimension is required to match the asymmetric shape of the high power oscillations in monocrystalline or ceramic PMN. The dependence of oscillations on sample thickness and spot size was qualitatively consistent with that observed in thermal defocussing materials. The temperature dependence of the oscillations was consistent with a previous numerical calculation based on the variation in the dn/dT with temperature, which has a broad maximum around 30 – 40°C.

In the shape of the temporal oscillations, monocrystalline PMN is intermediate between ceramic PMN and PLZT. As Krumins has pointed out³, the thermal conductivity K_T of monocrystalline PMN (10 W/mK) is much higher than for ceramic PLZT (1.0 W/mK) or ceramic PMN (0.08 W/mK) but the effective absorption coefficient α should be much lower than for either of the ceramics, due to the absence of scattering at grain boundaries or voids. Since thermal lens power is proportional to α/K_T , this is consistent with the higher power densities observed for thermal lensing and oscillation.

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