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Optical bistability in periodically poled LiNbO₃ induced by cascaded second-order non-linearity and the electro-optic effect

Yi-qiang Qin, Yong-yuan Zhu, Shi-ning Zhu and Nai-ben Ming

Department of Physics, National Laboratory of Solid State Microstructures, Nanjing University and Centre for Advanced Studies in Science and Technology of Microstructures, Nanjing 210093, People's Republic of China

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Abstract. We report our theoretical studies on optical bistability in a periodically poled LiNbO₃ crystal. The optical resonance and the non-linearity essential for bistability are induced by the electro-optic effect and the cascaded $\chi^{(2)}:\chi^{(2)}$ effect, respectively. It is found that the threshold for bistability is much lower than those based on the traditional periodically layered structure.

1. Introduction

Optical non-linearity and bistability have been topics of great interest for the past two decades because of their potential applications. It has been shown, both theoretically [1–3] and experimentally [4–7], that optical bistability can occur in one-dimensional (1-D) periodic media with Kerr-type non-linearity. In these traditional periodic media, each period consists of two layers: one layer with a constant refractive index and another one with an intensity-dependent refractive index. These 1-D periodic structures are always heterogeneous, and they are termed here two-component superlattices.

On the other hand, the cascaded second-order non-linearity [8, 9] can be used to produce an equivalent Kerr-like non-linearity, which appears in the form $\chi^{(2)}:\chi^{(2)}$. Here $\chi^{(2)}$ is the second-order non-linearity susceptibility tensor. The $\chi^{(2)}:\chi^{(2)}$ process occurs whenever waves interact parametrically via $\chi^{(2)}$, exchanging power and producing a Kerr-like non-linearity. Up- and down-conversion is necessary for the non-linearity to occur (figure 1). Recently, a large number of phenomena arising from cascaded second-order non-linear-optical processes, such as squeezed-light generation [10, 11], phase conjugation [12], large non-linear phase shifts [13], all-optical switching [14], soliton excitation [15], pulse shortening [16], and optical transistors [17], have been reported. These non-linear-optical phenomena can occur in various structures. Among them, a particular type of periodic structure has attracted much attention; it is composed of one ferroelectric material but with its spontaneous polarization direction modulated periodically. That is, the structure is homogeneous. In a ferroelectric material such as LiNbO₃ (LN) or LiTaO₃ (LT), the domain structure is of the 180° type. The refractive index is constant throughout the sample, which limits the production of an optical resonance. However, this situation can be changed by an external electric field through the electro-optic effect because of the sign reversal of the electro-optic coefficient in antiparallel domains. Therefore this homogeneous superlattice

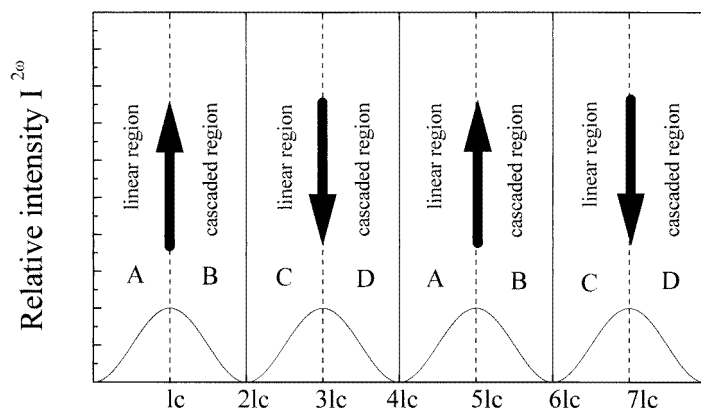


Figure 1. A schematic diagram of periodically poled LiNbO₃ with four components. The refractive indexes of the individual components are $n_0 + \delta n$ (A regions), $n_0 + \delta n + (6\pi/(n_0 + \delta n))\chi^{casc}|E(x)|^2$ (B regions), $n_0 - \delta n$ (C regions) and $n_0 - \delta n + (6\pi/(n_0 - \delta n))\chi^{casc}|E(x)|^2$ (D regions), respectively. The curve shows the dependence of the second-harmonic intensity on distance in a non-linear crystal with non-phase-matched interaction.

structure is advantageous for modulation of the refractive index compared to the traditional heterogeneous structure obtained by growth techniques such as metallo-organic chemical vapour deposition and pulsed laser deposition, especially for samples with a large number of periods. A thick sample needs excessive growth time and makes it difficult to maintain a uniform periodicity along the growth axis. As was shown above, the homogeneous superlattice structure has the following two characteristics: an intensity-dependent refractive index change and an optical resonance effect, which are required for the creation of optical bistability.

In this paper some theoretical studies have been made of this kind of structure, taking LN as an example. It is found that, under certain conditions, optical bistability will occur, and the threshold for the onset of bistability is lower than that of the traditional periodic structure by one to two orders of magnitude. In section 2, a physical model for optical bistability in the superlattice is given. Section 2 also includes an elaboration of the computational formalism. The optimum parameters for a periodically domain-inverted sample of LN obtained from QPM theory, and the computational results, are presented in section 3. We also conclude with brief comments in this section.

2. The mechanism and method

Field-induced periodic domain reversal can be realized in a bulk non-linear material such as LN or LT [18–20]. Usually, the domains are arranged periodically along the x -axis. According to QPM theory, in order to obtain cascaded $\chi^{(2)}:\chi^{(2)}$, the widths of the positive domains and the negative domains must be larger than one coherence length. For simplicity, in this paper the domain width is chosen to be double the coherence length. Inside each domain, in the region $0-l_c$, the fundamental field drives a second-harmonic field. In the region l_c-2l_c , the second harmonic will be down-converted to a fundamental because of phase mismatch; this pattern is repeated throughout the crystal. This situation is shown in figure 1. The coherence length l_c equals $\lambda/[4(n_{2\omega} - n_\omega)]$, where λ is the fundamental wavelength in vacuum, and n_ω and $n_{2\omega}$ are the refractive indices for the fundamental and

the second harmonic, respectively.

The cascaded process provides a new contribution to the non-linear index of refraction. In terms of the theory of SHG, the cascaded second-order non-linearity can be approximated as [8, 21]

$$\chi^{casc} = -\frac{|\chi^{(2)}|^2}{\Delta k} \frac{16\pi\omega}{n_{2\omega}c} \quad (1)$$

where $\Delta k = k^{2\omega} - 2k^\omega$, and ω and k^ω are the angular frequency and wavenumber of the fundamental beam, respectively; $k^{2\omega}$ is the wavenumber of the second harmonic, c is the speed of light in vacuum, $\chi^{(2)}$ is the second-order non-linearity. This Kerr-like non-linearity should be taken into account when an intense laser beam is used.

The sign and the magnitude of the non-linear response due to the cascaded process can be adjusted by phase mismatch: $k^{2\omega} - 2k^\omega = 4\pi(n_{2\omega} - n_\omega)/\lambda$. In our case, χ^{casc} is negative because the phase mismatch is positive due to the normal dispersion. When the laser beam incident onto the sample is weak, the cascaded effect can be neglected. In this case, under the action of an external electric field, the structure is just like that of a two-component superlattice. Because the antiparallel domains correspond to a sign reversal of the electro-optic coefficient, with an external electric field applied along the z -axis of the sample, the refractive index can be modulated from n_0 to $n_0 + \delta n$ (positive domains) or to $n_0 - \delta n$ (negative domains) where n_0 is the refractive index of a bulk material. However, if the laser beam is intense, the situation becomes quite different. The cascaded effect should be taken into consideration. Now each domain (positive or negative) can be divided into two parts: a linear dielectric region (such as $0-l_c$) with the refractive index $n_0 \pm \delta n$ and a non-linear region (such as l_c-2l_c) caused by cascaded $\chi^{(2)}:\chi^{(2)}$ with the refractive index $n_0 \pm \delta n + (6\pi/n_\omega)\chi^{casc}|E_1(x)|^2$ [4], where $E_1(x)$ is the fundamental field at point x and n_ω equals $n_0 \pm \delta n$. Thus the superlattice can be viewed as composed of four components, as is also shown in figure 1. That is, with the cascaded effect, the composition of the superlattice changes from having two components to having four components. Here, the usual third-order susceptibility term is neglected. The reason for this is as follows. In each domain, the usual third-order susceptibility exists inside both of the regions $0-l_c$ and l_c-2l_c , and the magnitude of the fundamental light varies continuously, therefore making no contribution to the transformation from a two-component structure to a four-component one. In our case, however, there is no cascaded effect inside the regions $0-l_c$, whereas inside the regions l_c-2l_c , the cascaded effect changes the refractive index of the superlattice. Also, due to the largest non-linear coefficient d_{33} of LN being used, χ^{casc} ($\sim 10^{-11}$ cm² W⁻¹) is larger than the usual third-order susceptibility ($\sim 10^{-12}$ cm² W⁻¹) by one order of magnitude [8, 22]. The problem of calculating the transmission through the structure can be solved numerically [2, 23] by the use of the appropriate boundary conditions and the transfer-matrix method [1, 4, 24]. In this paper, we have generalized the transfer-matrix method in which the non-linear regions are divided into a stack of very thin films.

So let us examine the case where the bulk material contains M positive and negative domains. $0 \leq m \leq M$ is the space occupied by the domains. However, in the non-linear regions, the values of $n = n_0 \pm \delta n + (6\pi/n_\omega)\chi^{casc}|E_1(x)|^2$ are not constant everywhere, because of the intensity-dependent refractive index. To overcome this difficulty, we divide each non-linear region into a large number of sublayers whose thicknesses are very small compared to the wavelength, so that the intensity and the non-linear coefficient in each sublayer can be considered constant. If the intensity-dependent refractive index of each sublayer is determined, the transfer-matrix method can be employed in the same way as in the case of linear media. The bulk material is impinged upon by a wave from the left,

in the region $x < x_0$, and then by one from the right. Where $x > x_M$, we have only the transmitted wave. We may write down the single Cartesian component of the electric field of the laser beam in the form

$$E(x, t) = E(x) \exp(i\omega t)$$

over the space of the domains.

$$E_{m,n} = \begin{cases} S \exp(ik_1 x) + R \exp(-ik_1 x) & x < x_0 \\ F_{m,n} \exp[ik_{m,n}(x - x_{m,n})] \\ \quad + B_{m,n} \exp[-ik_{m,n}(x - x_{m,n})] & x_{m,n} < x < x_{m,n+1} \\ T \exp[ik_1(x - x_M)] & x \geq x_M. \end{cases} \quad (2)$$

Here $E_{m,n}(x)$ is the field within the n th sublayer of the m th domain (in the non-linear medium). The corresponding wave vector is $k_{m,n}$ ($k_{m,n} = 2\pi n_{m,n}(x)/\lambda$). In the linear regions, $k_1 = 2\pi(n_0 \pm \delta n)/\lambda$.

According to the boundary conditions that require the continuity of the tangential components of \mathbf{E} and its derivative, we can obtain the recurrence relations corresponding to the linear region and the non-linear region:

$$\begin{aligned} F_m &= \frac{1}{2} \exp(-ik_1 l_c) \left[1 + \frac{n_{m,0}}{n_1} \right] F_{m,0} + \left[1 - \frac{n_{m,0}}{n_1} \right] B_{m,0} \\ B_m &= \frac{1}{2} \exp(+ik_1 l_c) \left[1 - \frac{n_{m,0}}{n_1} \right] F_{m,0} + \left[1 + \frac{n_{m,0}}{n_1} \right] B_{m,0} \end{aligned} \quad (3)$$

$$n_{m,0} = n_0 \pm \delta n + (6\pi/n_\omega) \chi^{casc} |F_{m,0} + B_{m,0}|^2$$

and

$$\begin{aligned} F_{m,n-1} &= \frac{1}{2} \exp(-ik_{m,n-1} d^*) \left[1 + \frac{n_{m,n}}{n_{m,n-1}} \right] F_{m,n} + \left[1 - \frac{n_{m,n}}{n_{m,n-1}} \right] B_{m,n} \\ B_{m,n-1} &= \frac{1}{2} \exp(+ik_{m,n-1} d^*) \left[1 - \frac{n_{m,n}}{n_{m,n-1}} \right] F_{m,n} + \left[1 + \frac{n_{m,n}}{n_{m,n-1}} \right] B_{m,n} \end{aligned} \quad (4)$$

$$n_{m,n} = n_0 \pm \delta n + (6\pi/n_\omega) \chi^{casc} |F_{m,n} \exp(ik_{m,n} d^*) + B_{m,n} \exp(-ik_{m,n} d^*)|^2$$

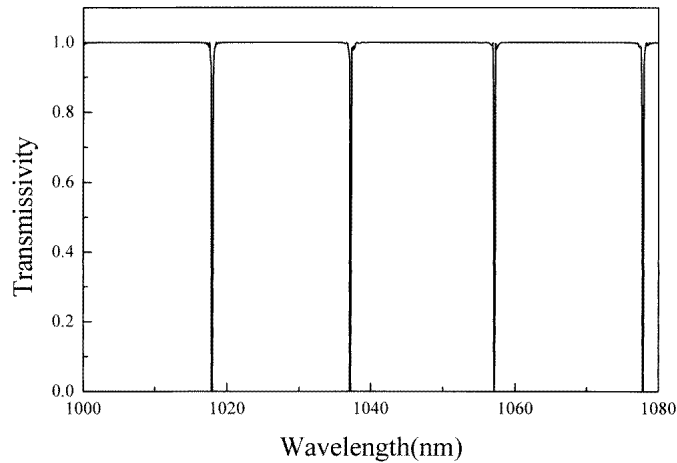
$$n_{m,n-1} = n_0 \pm \delta n + (6\pi/n_\omega) \chi^{casc} |F_{m,n} + B_{m,n}|^2$$

where F and B with one subscript are for the linear region ($0-l_c$) and with two subscripts, m, n , are for the non-linear region (l_c-2l_c). d^* is the width of the non-linear sublayer ($d^* = l_c/N$). The formulae are suitable for both positive domains and negative domains.

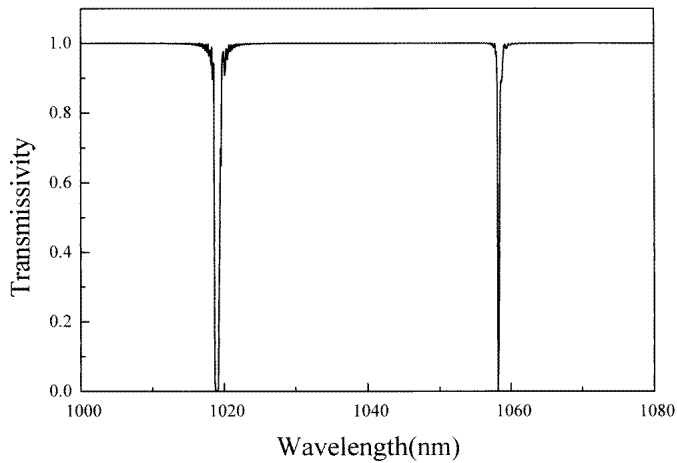
We use a double iterative method (m, n) to solve the hierarchy of equations given as equations (3) and (4). The above analysis is based on a plane-wave model. Usually Gaussian beams are used. Therefore it is necessary to investigate the validity of the plane-wave analysis for the non-linear process. According to references [25], if the magnitude of the sample length L is much smaller than the confocal parameter, the beam cross section remains essentially a constant within the sample and the plane-wave analysis can be used.

3. Results and discussion

The linear and non-linear transmissivity spectra calculated with the above method for a periodically poled sample of LN are shown in figure 2. In our case, $\chi^{casc} \sim 10^{-11} \text{ cm}^2 \text{ W}^{-1}$ and the domain width equals $6.6 \mu\text{m}$ which is much larger than the wavelength. Figure 2(a) shows the linear case, which corresponds to weak incident intensity. There are periodic



(a)



(b)

Figure 2. The transmissivity obtained as a function of the wavelength for an index-modulated LiNbO₃ sample utilizing the electro-optic effect. The width of the domains equals 6.6 μm . $n_0 = 2.1562$, $\delta n = 0.002$, corresponding to an external electric field of the order of 10^4 V mm^{-1} . (a) The linear response with the refractive indexes $n_0 + \delta n$ (positive domains) and $n_0 - \delta n$ (negative domains). (b) The non-linear response with refractive indexes $n_0 \pm \delta n$ (linear regions) and $n_0 \pm \delta n + (6\pi/n_\omega)\chi^{casc}|E(x)|^2$ (non-linear regions), with $n_\omega \pm n_0 + \delta n$, $\chi^{casc} \sim 10^{-11} \text{ cm}^2 \text{ W}^{-1}$, where M is the number of domains and each domain is divided into N sublayers. $M = 1000$ and $N = 200$.

optical stop bands as well as a series of allowed bands. The transmissivity spectrum exhibits sharp gradients in the vicinity of the forbidden band where the resonances are closely spaced. This fact is useful for producing bistability. Figure 2(b) shows the case in which the refractive index in the non-linear regions is dependent on the light intensity caused by cascaded second-order non-linearity. The non-linear spectrum also exhibits the periodicity of the forbidden band. The location and shape of the forbidden band depends strongly on the modulation of the refractive index due to the electro-optic effect and the

cascaded non-linear effect. When the strength of the applied external electric field increases, the difference between the refractive indexes of the positive and negative domains increases, which leads to the enhancement of the resonance effect and the increase in the number of forbidden bands. Comparing with figure 2(a), the period of the non-linear spectrum shown in figure 2(b) is seen to be almost double. The reason for this may be the conversion of a two-component structure into a four-component structure. We optimized the parameters such as the index modulation and the domain width in order to ensure that the operating wavelength was at the edge of the forbidden band and that the corresponding domain width was double the coherence length. The details of the optimized parameters are noted in the caption of figure 2. $\delta n = 0.002$ corresponds to an external electric field of the order of 10^4 V mm^{-1} . Moreover, in references [2], the authors found that for $\chi^{(3)} < 0$, the bistability occurs near the lower gap edge, while for $\chi^{(3)} > 0$, the bistability occurs near the upper gap edge. Here, in our case, the bistability is exhibited at the lower gap edge because of the negative cascaded non-linearity.

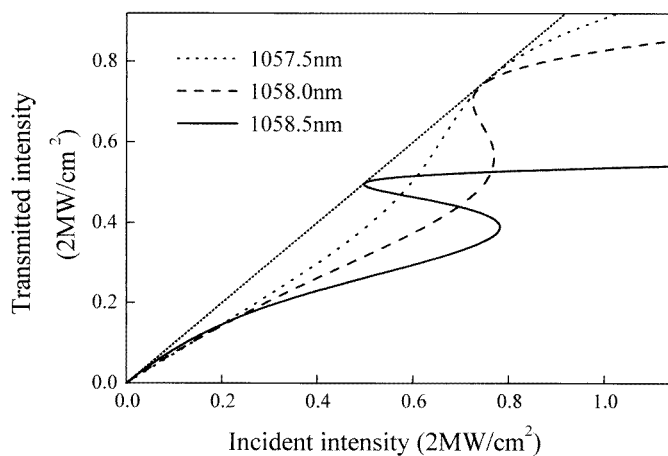


Figure 3. The transmitted intensity as a function of the incident intensity calculated for three different wavelengths with the same superlattice parameters as in figure 2. The unit of the incident and transmitted intensities is 2 MW cm^{-2} . The dashed curve corresponds to the case with a transmissivity of unity.

A non-linear periodic layered structure can exhibit optical differential gain and hysteresis in its response to an input intensity. Figure 3 shows the transmitted output intensity as a function of the input intensity, calculated for three different wavelengths. The linear forbidden band is at 1057.0 nm. The detunings corresponding to three wavelengths are 0.5, 1.0 and 1.5 nm, respectively. When the detuning is larger than a certain critical value, the output–input function exhibits bistable hysteresis. The unit of the incident and transmitted intensities is 2 MW cm^{-2} . It should be noted here that the occurrence of bistability only requires a refractive index variation of the order of 10^{-5} . This is much lower than for the two-component structure (10^{-3} – 10^{-4}) [26]. We have also calculated the threshold for the onset of bistability in the two-component structure with the same parameters, and found that the threshold is higher by one to two orders of magnitude. The reasons for this may be that the four-component structure produces a double-resonance effect and that the non-linear refractive index has a sharp spatial variation in the periodically poled LN structure due to the cascaded non-linearity.

In summary, a periodically poled LN structure is proposed for optical bistability operation. The resonance and the non-linearity necessary for the bistability are induced by the electro-optic effect and the cascaded non-linear effect, respectively. Using a generalized transfer-matrix method, we have shown that the optical bistability occurs under certain conditions. The threshold for bistability can be lower than that for a usual periodic structure.

Acknowledgments

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