Nonlinear Magneto-Optical Bragg Gratings

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In magneto-optical gratings the magnetic dipolar coupling superimposed on the electric dipolar one introduces nonreciprocity and spectral discrimination between circular polarization states, measured by a Zeeman-like splitting of the photonic Bragg resonances. In a nonlinear regime the degree of nonreciprocity is modified by the photoinduced interplay of these splittings and their Stark-like shifts. We predict novel magneto-optical modulation schemes for switching between orthogonal circular polarization states of transmission or reflection operated by means of an intense linearly polarized optical pulse train.

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In recent years a certain activity has been manifested in the control of optical beams with the use of photonic gratings [1], by interfacing optical materials and modulating the dielectric constant over characteristic scales of the order of the optical wavelength. The main issue here is the redistribution and tuning of the density of states of the electromagnetic field and concomitantly the spectral features of the excitations that the grating can sustain; these features can also be affected by photoinduced changes of the dielectric constant in the nonlinear regime [2,3]. Although not explicitly stated in these cases time reversal symmetry is implicitly assumed and this, in particular, imposes the wave propagation being reciprocal. In this respect the situation radically changes as a modulated magnetic induction is superimposed with a period commensurate to that of the grating. In such magneto-optical gratings the time reversal symmetry breaks down and entails the system with nonreciprocity [4]; this lifts the left (LCP) and right (RCP) circular polarization state degeneracy and introduces a distinction between forward and backward propagation. This profoundly affects the polarization state evolution of the beams since the resonance conditions of the counterpropagating beams are strongly affected as is also their nonlinear coupling.

In this Letter we show that the interplay between nonlinearity and nonreciprocity leads to novel operation schemes for polarization state modulation and switching in a magneto-optical binary Bragg grating. Restricting ourselves to the case of one-dimensional gratings formed by piling up alternating magnetic and nonmagnetic layers with the light incident normally on this stack there are several possible configurations differing by the order the piling up is made and the insertion of intentional gyrotropic defects.

For our purpose it is sufficient to consider the simple magneto-optical grating with a single defect as depicted in Fig. 1: a magnetic layer of thickness $d_{N/2}$ is sandwiched between two identical periodic gratings formed by a repeat unit consisting of a pair of a magnetic and a nonmagnetic layer both assumed lossless; the introduced center defect effectively acts as a local phase shift of the otherwise periodic structure, opening for a narrow transmission window in the optical band gap [1,2]. For simplicity we consider the case of an identical number of repeat units (periods) on either side of the defect; other configurations can, however, be treated analogously without much additional complication.

The fabrication of such gratings has been recently reported [5–7] by piling up double layers of garnets such as Bi:YIG/GaGdG or Bi:YIG/SiO_2, with the gyrotropic defect being, for instance, a thin layer of a magnetic metal Fe, Ni, Co, or other magnetic compound [4]. To fix the ideas we use parameters pertinent to such gratings, and choose the layer thicknesses to correspond to a quarter wavelength so as to work in the transmission mode. One can also exploit the outstanding performances achieved in molecular beam epitaxial growth of layers of semimagnetic semiconductors such as $Cd_{1-x}Mn_xTe$, with the defect being a quantum well with prescribed spectral characteristics. In such structures giant photoinduced Faraday and magneto-optic Kerr rotation has been evidenced [8,9] in the nonlinear regime; in such structures one must, however, work

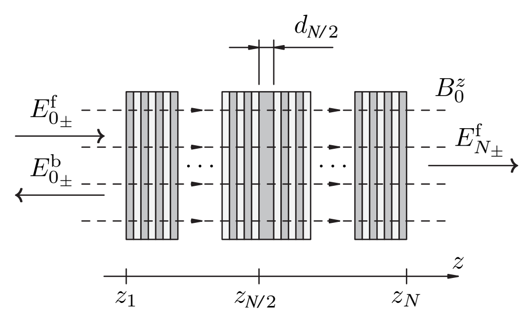

FIG. 1. The nonlinear magneto-optical crystal in the Faraday configuration. The setup consist of $N-1$ layers of alternating optical and magneto-optical properties, with the centermost layer possessing twice the thickness of other layers with identical material properties.

close to exciton polaritonic resonances, which introduce resonant enhancement but also absorption losses and resonance dynamics which complicates the following discussion in the dispersive regime.

First, we outline the algorithm of calculation as used in the analysis of the transmission properties of the magneto-optical structure, restricting to the case of plane waves. In each homogeneous layer $z_k < z < z_{k+1}$, $k = 1, 2, \ldots N-1$, the components of the plane-parallel electromagnetic field $\mathbf{E}(z, t) = \mathrm{Re}[\mathbf{E}_\omega \exp(-i\omega t)]$ propagate collinearly with an externally applied static magnetic field $\mathbf{B}_0 = B_0^z \mathbf{e}_z$, and the local constitutive relation for the electric polarization density $\mathbf{P}(z, t) = \mathrm{Re}[\mathbf{P}_\omega \exp(-i\omega t)]$ of the medium is [10]

$$\begin{aligned}\mathbf{P}_\omega = \varepsilon_0[&\chi^{\mathrm{ee}}_{xx}\mathbf{E}_\omega + \chi^{\mathrm{eem}}_{xyz}\mathbf{E}_\omega \times \mathbf{B}_0 + \tfrac{3}{4}(\chi^{\mathrm{eeee}}_{xxxx} - \chi^{\mathrm{eeee}}_{xyyx})\\ &\times (\mathbf{E}_\omega \cdot \mathbf{E}^*_\omega)\mathbf{E}_\omega + \tfrac{3}{4}\chi^{\mathrm{eeee}}_{xyyx}(\mathbf{E}_\omega \cdot \mathbf{E}_\omega)\mathbf{E}^*_\omega\\ &+ \tfrac{3}{4}\chi^{\mathrm{eeeem}}_{xyyyz}(\mathbf{E}_\omega \cdot \mathbf{E}^*_\omega)\mathbf{E}_\omega \times \mathbf{B}_0\\ &+ \tfrac{3}{4}\chi^{\mathrm{eeeem}}_{xxxyz}\mathbf{E}_\omega(\mathbf{E}_\omega \cdot (\mathbf{E}^*_\omega \times \mathbf{B}_0))].\end{aligned}$$

In this expression, the spatial dependence of the fields and susceptibilities is omitted in notation, as is also any slow temporal variation of the fields; from now on each appearance of a field or susceptibility implicitly implies evaluation in respective layer of context. In each layer, the envelope of the electromagnetic field satisfies the autonomous nonlinear wave equation

$$\frac{\partial^2 \mathbf{E}_\omega}{\partial z^2} + (\omega/c)^2 \mathbf{E}_\omega = -\mu_0 \omega^2 \mathbf{P}_\omega. \tag{1}$$

The field is in each layer expressed in a circularly polarized basis $\mathbf{e}_\pm = (\mathbf{e}_x \pm i\mathbf{e}_y)/\sqrt{2}$, with forward and backward traveling modes as a superposition

$$\begin{aligned}\mathbf{E}_\omega = &[\mathbf{e}_+ E^{\mathrm{f}}_{k_+}(z) + \mathbf{e}_- E^{\mathrm{f}}_{k_-}(z)]\exp[i\omega n_k(z - z_k)/c]\\ &+ [\mathbf{e}^*_+ E^{\mathrm{b}}_{k_+}(z) + \mathbf{e}^*_- E^{\mathrm{b}}_{k_-}(z)]\exp[-i\omega n_k(z - z_k)/c],\end{aligned}$$

where $n_k = (1 + \chi^{\mathrm{ee}}_{xx})^{1/2}$ are the linear refractive indices of the layers.

By applying the slowly varying envelope approximation to Eq. (1), the general solutions for the field components in lossless media become [10]

$$E^{\mathrm{f}}_{k_\pm} = A^{\mathrm{f}}_{k_\pm} \exp[i(\omega/c)(n^{\mathrm{f}}_{k_\pm} \pm g_k)(z - z_k) + i\psi^{\mathrm{f}}_{k_\pm}], \tag{2a}$$

$$E^{\mathrm{b}}_{k_\pm} = A^{\mathrm{b}}_{k_\pm} \exp[-i(\omega/c)(n^{\mathrm{b}}_{k_\pm} \mp g_k)(z - z_k) + i\psi^{\mathrm{b}}_{k_\pm}]. \tag{2b}$$

In these expressions $A^{\mathrm{f,b}}_{k_\pm}$ and $\psi^{\mathrm{f,b}}_{k_\pm}$ are real constants of integration, determined by the boundary conditions of the kth layer, and $g_k = i\chi^{\mathrm{eem}}_{xyz}B_0^z/(2n_k)$ are the real linear magneto-optical gyration coefficients. In Eqs. (2) the nonlinear optical and magneto-optical light-matter interactions are described by the field-dependent parameters

$$\begin{aligned}\eta^{\mathrm{f}}_{k_\pm} &= p_{k_\pm}(A^{\mathrm{f}2}_{k_\pm} + 2A^{\mathrm{b}2}_{k_\mp}) + q_{k_\pm}(A^{\mathrm{f}2}_{k_\mp} + A^{\mathrm{b}2}_{k_\pm}),\\ \eta^{\mathrm{b}}_{k_\pm} &= p_{k_\mp}(A^{\mathrm{b}2}_{k_\pm} + 2A^{\mathrm{f}2}_{k_\mp}) + q_{k_\mp}(A^{\mathrm{b}2}_{k_\mp} + A^{\mathrm{f}2}_{k_\pm}),\end{aligned}$$

where the coefficients $p_{k_\pm}$ and $q_{k_\pm}$ in the nonlinear susceptibility formalism [11] are expressed as

$$\begin{aligned}p_{k_\pm} &= \frac{3}{8n_k}[\chi^{\mathrm{eeee}}_{xxxx} - \chi^{\mathrm{eeee}}_{xyyx} \pm i(\chi^{\mathrm{eeeem}}_{xyyyz} - \chi^{\mathrm{eeeem}}_{xxxyz})B_0^z],\\ q_{k_\pm} &= \frac{3}{8n_k}[\chi^{\mathrm{eeee}}_{xxxx} + \chi^{\mathrm{eeee}}_{xyyx} \pm i(\chi^{\mathrm{eeeem}}_{xyyyz} + \chi^{\mathrm{eeeem}}_{xxxyz})B_0^z].\end{aligned}$$

The angle of rotation ϑ_k of the polarization ellipse of the forward traveling components around the z axis in each layer is then

$$\begin{aligned}\vartheta_k = \frac{\omega(z - z_k)}{2n_k c}[&-i\chi^{\mathrm{eem}}_{xyz}B_0^z + (3/4)\chi^{\mathrm{eeee}}_{xyyx}(A^{\mathrm{f}2}_{k_+} - A^{\mathrm{f}2}_{k_-})\\ &- (3/4)i\chi^{\mathrm{eeeem}}_{xyyyz}(A^{\mathrm{f}2}_{k_+} + A^{\mathrm{f}2}_{k_-})B_0^z\\ &+ (3/8)(\chi^{\mathrm{eeee}}_{xxxx} - 3\chi^{\mathrm{eeee}}_{xyyx})(A^{\mathrm{b}2}_{k_+} - A^{\mathrm{b}2}_{k_-})\\ &+ (3/8)i(\chi^{\mathrm{eeeem}}_{xxxyz} - 3\chi^{\mathrm{eeeem}}_{xyyyz})(A^{\mathrm{b}2}_{k_+} + A^{\mathrm{b}2}_{k_-})B_0^z].\end{aligned}$$

The first term in this expression describes linear Faraday rotation [4] and leads to an effective Zeeman-like polarization state splitting of the doubly degenerate Bragg resonances. The second and fourth terms arise from optical Kerr effect and lead to photoinduced modification of the ellipse rotation for forward and backward propagating waves, respectively. Referring to Eq. (1) these terms effectively act to give photoinduced Stark-like shifts of the split Bragg resonances. Finally the third and fifth terms describe photoinduced magneto-optic Faraday rotation or equivalently photoinduced modification of the effective Zeeman-like splitting of the Bragg resonances. We note in passing that the fourth and fifth terms vanish whenever Kleinman symmetry [11] holds.

By neglecting nonlinear effects at the discrete interfaces between the homogeneous layers, the continuity requirement of the transverse electromagnetic field is formulated by the boundary conditions

$$E^{\mathrm{f}}_{k_\pm}(z_k) = \tau_{k_\pm}E^{\mathrm{f}}_{k-1_\pm}(z_k)\exp(i\omega n_{k-1}d_{k-1}/c) + \rho'_{k_\mp}E^{\mathrm{b}}_{k_\mp}(z_k), \tag{3a}$$

$$E^{\mathrm{b}}_{k_\mp}(z_{k+1})\exp(-i\omega n_k d_k/c) = \tau'_{k+1_\mp}E^{\mathrm{b}}_{k+1_\mp}(z_{k+1}) + \rho_{k+1_\pm}E^{\mathrm{f}}_{k_\pm}(z_{k+1})\exp(i\omega n_k d_k/c), \tag{3b}$$

for $k = 1, 2, \ldots, N-1$, where $d_k = z_{k+1} - z_k$ are the layer thicknesses, and where

$$\rho_{k_\pm} = \frac{n_{k-1} - n_k \pm (g_{k-1} - g_k)}{n_{k-1} + n_k \pm (g_{k-1} + g_k)} = -\rho'_{k_\mp}, \qquad \tau_{k_\pm} = \frac{2(n_{k-1} \pm g_{k-1})}{n_{k-1} + n_k \pm (g_{k-1} + g_k)} = (1 - \rho^2_{k_\pm})/\tau'_{k_\mp}$$

are the reflection and transmission coefficients at the interfaces, including linear magneto-optical effects. In order to solve Eqs. (2) and (3) for the relation between incident ($k = 0$) and transmitted ($k = N$) fields, we impose the additional boundary condition that the backward propagating field is zero at the exit end of the grating. By iteratively using Eqs. (3) and (2) in the order $k = N-1, N-2, \ldots, 1$, consecutively solving for lower-indexed electric fields, we obtain an algorithm for solving the inverse problem, calculating the incident and reflected optical fields corresponding to a transmitted optical field.

The evolution of the optical field is conveniently expressed in terms of the Stokes parameters, which for the input light are taken as

$$S_0 = |E^{\mathrm{f}}_{0_+}|^2 + |E^{\mathrm{f}}_{0_-}|^2, \qquad S_1 = 2\,\mathrm{Re}[E^{\mathrm{f}*}_{0_+} E^{\mathrm{f}}_{0_-}],$$
$$S_3 = |E^{\mathrm{f}}_{0_+}|^2 - |E^{\mathrm{f}}_{0_-}|^2, \qquad S_2 = 2\,\mathrm{Im}[E^{\mathrm{f}*}_{0_+} E^{\mathrm{f}}_{0_-}].$$

The parameter S_0 is a measure of the intensity of the wave, and S_3 a measure of the ellipticity of the polarization state, while the parameters S_1 and S_2 indicate the amount of power contained in the x and y directions, serving to determine the orientation of the polarization ellipse. Similarly, the Stokes parameters for the transmitted light are taken as

$$W_0 = |E^{\mathrm{f}}_{N_+}|^2 + |E^{\mathrm{f}}_{N_-}|^2, \qquad W_1 = 2\,\mathrm{Re}[E^{\mathrm{f}*}_{N_+} E^{\mathrm{f}}_{N_-}],$$
$$W_3 = |E^{\mathrm{f}}_{N_+}|^2 - |E^{\mathrm{f}}_{N_-}|^2, \qquad W_2 = 2\,\mathrm{Im}[E^{\mathrm{f}*}_{N_+} E^{\mathrm{f}}_{N_-}].$$

In terms of S_0 and W_0, the incident and transmitted intensities in SI units are $I_{\mathrm{in}} = \varepsilon_0 c S_0/2$ and $I_{\mathrm{tr}} = \varepsilon_0 c W_0/2$.

The described algorithm of calculation can be applied to a wide range of magneto-optical gratings, singly or multiply periodic, chirped, or even random without appreciable complication. Here we apply it to analyze the transmission behavior of the binary nonlinear magneto-optical grating with one gyrotropic defect layer inserted, to show the principle of a novel scheme of polarization state modulation and switching. To illustrate the principle we use a structure consisting of a moderate and odd number of $N - 1 = 53$ layers of alternating optical and magneto-optical coefficients, designed to be resonant at a center vacuum wavelength of $\lambda_{\mathrm{c}} = 1150$ nm in the absence of static magnetic fields and strong optical fields. The layer thicknesses are chosen as $d_k = \lambda_{\mathrm{c}}/4n_k$, providing a broad-banded reflection around the center wavelength; however, in order to open a transmission peak in the band gap of the structure, the centermost layer ($k = N/2$) is taken as an intentionally introduced defect layer [1,2] of thickness $d_{N/2} = \lambda_{\mathrm{c}}/2n_{\mathrm{o}}$, but otherwise with the same material properties as all other odd layers. The elements of the chosen material tensors are listed in Table I.

Because of their nonlinear and multivalued nature, the precise transmission and reflection characteristics of this magneto-optical structure must be calculated using the outlined inverse algorithm; however, the principle of operation can be qualitatively explained in terms of the interplay of the Zeeman-like splitting and photoinduced Stark-like shifts of the eigenmodes of the underlying photonic structure as will now be illustrated.

TABLE I. Material parameters used in numerical simulation.

Odd layers	Even layers	Unit
$\chi^{\mathrm{ee}}_{xx} = 3.8$ ($n_{\mathrm{o}} = 2.2$)	$\chi^{\mathrm{ee}}_{xx} = 2.6$ ($n_{\mathrm{e}} = 1.9$)	
$i\chi^{\mathrm{eem}}_{xyz} B^z_0 = 4.2 \times 10^{-2}$ [a]	$i\chi^{\mathrm{eem}}_{xyz} B^z_0 = 0$	
$\chi^{\mathrm{eeee}}_{xxxx} = 1.8 \times 10^{-18}$ [b]	$\chi^{\mathrm{eeee}}_{xxxx} = 0$	$\mathrm{m^2/V^2}$
$\chi^{\mathrm{eeee}}_{xyyx} = 0.36 \times 10^{-18}$ [c]	$i\chi^{\mathrm{eeee}}_{xyyx} = 0$	$\mathrm{m^2/V^2}$
$i\chi^{\mathrm{eeeem}}_{xyyyz} B^z_0 = 0.1 \times 10^{-18}$ [d]	$i\chi^{\mathrm{eeeem}}_{xyyyz} B^z_0 = 0$	$\mathrm{m^2/V^2}$
$i\chi^{\mathrm{eeeem}}_{xxxyz} B^z_0 = 0.3 \times 10^{-18}$	$i\chi^{\mathrm{eeeem}}_{xxxyz} B^z_0 = 0$	$\mathrm{m^2/V^2}$

[a]Corresponding to a linear Faraday rotation of 52 rad/mm, or $3.0°/\mu$m.
[b]Corresponding to a refractive index change of 2.3×10^{-3} for linearly polarized light at an intensity of 1 $\mathrm{GW/cm^2}$.
[c]Corresponding to a maximum nonlinear optical ellipse rotation of 2.5 rad/mm at 1 $\mathrm{GW/cm^2}$.
[d]Corresponding to a photoinduced Faraday rotation of 0.7 rad/mm at 1 $\mathrm{GW/cm^2}$ (1.3% of the linear rotation).

As the static magnetic field is applied on the structure, the degeneracy of resonance peaks of circularly polarized modes of the underlying photonic structure is lifted, due to the magneto-optically induced differential change in resonance conditions of the layers, and Zeeman-like doublets are set up. Such a transmission spectrum of the structure is in the linear regime depicted in Fig. 2. The separation of circularly polarized resonance peaks can be tuned to a spectral resolution $\Delta\lambda$ by means of the static magnetic field, and the polarization state selectivity in the resonant region can then be exploited by applying a dynamical modulation of the nonlinear characteristics of the grating. Because of the optical Kerr effect, a change of the refractive index of the layers can be modulated dynamically by an intense optical beam, resulting in Stark-like shifts of the

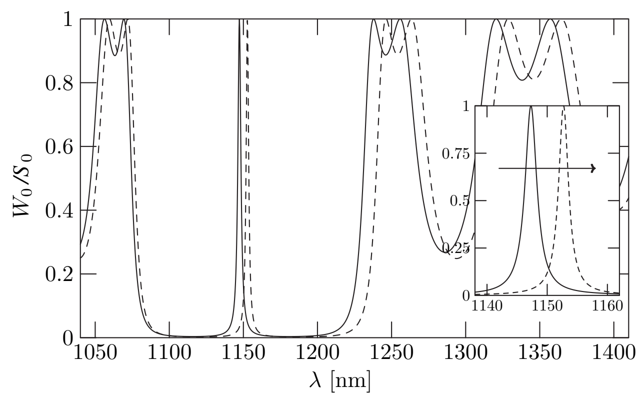

FIG. 2. Linear optical transmission for RCP (solid line) and LCP (dashed line) states of the incident optical wave vs vacuum wavelength $\lambda = 2\pi c/\omega$, for refractive index $n_{\mathrm{o}} = 2.18$ and gyration coefficient $g_{\mathrm{o}} = 9.6 \times 10^{-3}$ of all odd layers. The corresponding parameters for even layers are $n_{\mathrm{e}} = 1.93$ and $g_{\mathrm{e}} = 0$. The arrow in the inset graph shows the direction of displacement of the doublet of resonance peaks as the crystal is exposed to an intense light beam.

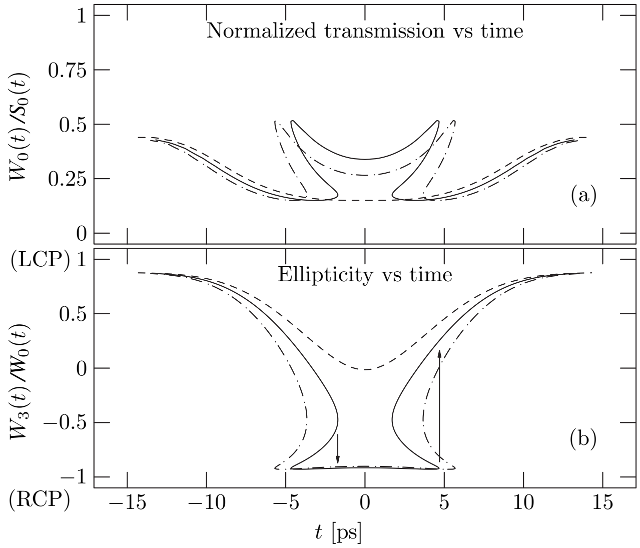

FIG. 3. The (a) normalized intensity transmission W_0/S_0 and (b) corresponding normalized ellipticity W_3/W_0 of the transmitted polarization state, as a function of time t for a linearly polarized input Gaussian pulse of half-maximum duration 10 ps and peak intensity 1.4 GW/cm^2 (dashed curves), 1.9 GW/cm^2 (solid curves), and 2.5 GW/cm^2 (dot-dashed curves). For a beam focused onto a spot of 40 μm diameter, the curves correspond to pulse energies of 190 nJ, 255 nJ, and 340 nJ, respectively.

resonance peaks of the doublets. In the following we assume positive index changes; however, the reverse situation is equally applicable. By operating the structure at the vacuum wavelength of the upper resonance peak, the peaks can be unidirectionally shifted in such a way that the structure instead becomes resonant for the orthogonal circular polarization. Employing an optical pulse train for modulation, we hence obtain means for dynamical switching between orthogonal circularly polarized modes of transmission. For this situation to arise clearly the peak intensity of the pulses should be tuned such that the photoinduced Stark-like shift matches the Zeeman-like splitting $\Delta\lambda$, as illustrated in Fig. 2.

The temporal transmission characteristics at vacuum wavelength $\lambda = 1152.7$ nm is in Fig. 3 illustrated for a Gaussian pulse $I_{\text{in}}(t) = I_{\text{peak}} \exp[-(2t/\Delta t)^2 \ln 2]$ of half-maximum duration $\Delta t = 10$ ps [12]. At low intensity, the structure is left circularly polarizing, while an increase of the peak intensity to $I_{\text{peak}} = 1.4$ GW/cm^2 causes a linearly polarized state to pass unchanged at $t = 0$. Increasing the peak intensity further, the transmission undergoes a bifurcation, effectively switching the structure to transmit right circular polarization states at high intensities. In particular, for a pulse train at $I_{\text{peak}} = 1.9$ GW/cm^2 the transmitted ellipticity is locked into a bistable modulation between LCP or RCP transmission, with the RCP state being reflected whenever the LCP state is transmitted and vice versa. The bistable polarization state switching cycle is in Fig. 3(b) indicated with arrows. By reversing the sign of the magnetic field, still with a linearly polarized input pulse train, the transmission is effectively inverted with $W_3 \to -W_3$.

It is noteworthy that in Fig. 3 the transmitted ellipticity in the switched state is flat and close to unity, hence providing a useful scheme for polarization state switching; this can be of interest in preparation of polarized photon states, in particular, for quantum information uses. In this respect it is also noteworthy that by mapping the Stokes parameters on the Poincaré sphere and also attaching the equation of motion (1) of the polarization state, a close analogy with the Maxwell-Bloch description of the evolution of two-level quantum system can be drawn.

In conclusion, we have in this Letter developed an approach to model the transmission and reflection characteristics of nonlinear magneto-optical gratings in terms of an algorithm for solving the inverse problem. In analogy with optical transitions we show that the gross features can be traced to the interplay of Zeeman-like splittings and Stark-like shifts of the eigenmodes of the underlying photonic structure. This introduces a coupling between polarization state and intensity that can be exploited to develop novel schemes of modulation between circularly polarized states of transmission or reflection. This and the bistable behavior are manifestations of the nonreciprocity that the presence of the static magnetic field entails these systems. Clearly the behavior can be tuned by means of the externally applied static magnetic field and the intensity of the light beam.

[1] K. Sakoda, *Optical Properties of Photonic Crystals* (Springer-Verlag, Berlin, 2001).
[2] *Nonlinear Photonic Crystals*, edited by R. E. Slusher and B. J. Eggleton (Springer-Verlag, Berlin, 2003).
[3] M. Soljačić and J. D. Joannopoulos, Nat. Mater. **3**, 211 (2004).
[4] A. K. Zvezdin and V. A. Kotov, *Modern Magnetooptics and Magnetooptical Materials* (Institute of Physics Publishing, London, 1997).
[5] H. Kato *et al.*, J. Appl. Phys. **93**, 3906 (2003).
[6] M. J. Steel, M. Levy, and R. M. Osgood, IEEE Photonics Technol. Lett. **12**, 1171 (2000).
[7] R. L. Espinola *et al.*, Opt. Lett. **29**, 941 (2004).
[8] C. Buss *et al.*, Phys. Rev. Lett. **78**, 4123 (1997).
[9] D. Pereda Cubian *et al.*, Phys. Rev. B **67**, 045308 (2003).
[10] F. Jonsson and C. Flytzanis, Phys. Rev. Lett. **82**, 1426 (1999).
[11] Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984).
[12] The Gaussian time-bandwidth product is $\Delta t \Delta\omega = 4 \ln 2$, where both the duration Δt and spectral width $\Delta\omega$ are given as full width at half maximum (FWHM) values. For a 10 ps pulse, the spectral width corresponds to a vacuum wavelength extent of 0.19 nm.