Bistability, multistability and non-reciprocal light propagation in Thue–Morse multilayered structures

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New Journal of Physics 12 (2010) 053041 (20pp)
Received 17 December 2009
Published 26 May 2010
Online at http://www.njp.org/
doi:10.1088/1367-2630/12/5/053041

Abstract. The nonlinear properties of quasi-periodic photonic crystals based on the Thue–Morse sequence are investigated. The intrinsic spatial asymmetry of these one-dimensional structures for odd generation numbers results in bistability thresholds, which are sensitive to the propagation direction. Along with resonances of perfect transmission, this feature allows us to achieve strongly non-reciprocal propagation and to create an all-optical diode. The salient qualitative features of such optical diode action are readily explained through a simple coupled resonator model. The efficiency of a passive scheme that does not necessitate an additional short pump signal is compared to an active scheme where such a signal is required.

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1. Introduction

The experimental discovery of quasicrystals [1] has given rise to a sudden breakthrough in the area of solid state physics. It had been assumed for a long time that a periodic arrangement of unit cells was the main prerequisite for the specific properties of crystalline matter. The structure of quasicrystals does exhibit long-range order or correlations between distant parts, but at the same time there is no underlying periodicity, in the sense that a shifted copy of the crystal never matches exactly the original one. In fact, quasicrystals represent an intermediate stage between random media and periodic crystals, effectively combining both localization properties as a result of short-range disorder and the presence of bandgaps due to long-range correlations [2].

The concept of quasi-periodicity was readily transferred to photonic crystals and proved to be of great value for many practical purposes [3–6]. Photonic quasicrystals are deterministically generated dielectric structures with a non-periodic modulation of the refractive index. In the one-dimensional (1D) case, they can be formed by stacking together dielectric layers of several different types according to the substitutional sequence under investigation (Cantor, Fibonacci, Rudin-Shapiro, Thue–Morse (ThM), etc) [7]. The Fibonacci sequence is of particular importance, since it leads to the existence of two incommensurable periods in the spatial spectrum of the structure. Such a behavior is typical of sequences with a so-called pure point spectrum, which makes the Fibonacci sequence truly quasi-periodic, as a consequence of the appearance of Bragg-like peaks in the spatial spectrum [8]. This property has been demonstrated to be very valuable for nonlinear optical applications, such as third harmonic generation [9]. In fact, the latter is a two-stage process, and for each stage, different phase matching conditions should be met. The Fibonacci sequence allows us to fulfill both of them simultaneously and on the same crystal [10].

In contrast to the Fibonacci sequence, the ThM sequence possesses a singular continuous spectrum, which is neither continuous nor singular [11, 12]. Thus, strictly speaking, the ThM sequence is not quasi-periodic, but rather deterministically aperiodic. To an even more different class belongs the Rudin–Shapiro sequence, which shows evidence of a continuous spectrum, analogous to the one exhibited by random sequences [13]. With a slight abuse of terminology,
we shall always refer to the above sequences as quasi-periodic, and we shall focus our attention specifically on the ThM sequence, for the reasons explained below.

The bandgaps of quasi-periodic multilayered structures, also called ‘pseudo bandgaps’ [14] or ‘fractal bandgaps’ in the literature [15], often contain resonant states, which can be considered as a manifestation of numerous defects distributed along the structure. Their quality factors are not very large when compared with those of symmetrically placed defects inside conventional Bragg structures, but the mode profiles associated with these resonances are extended in space and, as a consequence, are very suitable for the enhancement of nonlinear effects throughout the whole body of the crystal. In order to investigate the specific properties of bistability and multistability in quasicrystals, in the following we shall restrict our attention exclusively to 1D structures. As a representative case, we focus our attention on the ThM aperiodic multilayer. In making this choice we were guided by the following two reasons. Firstly, the spatial asymmetry of the ThM sequence (of odd generation numbers) interacts with the nonlinearity and makes the transmission sensitive to the propagation direction [16]. Secondly, almost all resonances in ThM quasicrystals are resonances of complete (i.e. 100%) transmission, with some of them being situated well inside the pseudo-bandgap regions. Taken together, the above two properties provide favorable conditions for the design of an all-optical diode, i.e. a device that shows a strong contrast in the transmission between forward and backward incidence. A proper understanding of bistability phenomena in quasi-periodic crystals is very important for the understanding of the optical diode action described above.

The paper is organized as follows. In section 2, we briefly review the well-known linear properties of ThM quasicrystals and classify their resonances by using the method of trace maps. It is shown that the field profiles at resonance frequencies follow the pattern of ThM sequence and the classification suggested can be used as a measure of this self-similarity. In section 3, we show that the spatial asymmetry inherent in ThM quasicrystals of odd generation numbers can interact with Kerr nonlinearity in such a way that transmission becomes sensitive to the propagation direction. It is very important that the dynamics of this interplay can have a sudden jump due to bistability when the intensity of the incident light changes, and the thresholds for such jumps, are different for the incidence from the left and from the right of the multilayer. In section 4, we apply a phenomenological model based on a coupled resonator model to explain this behavior analytically for both the bistable and multistable cases. The general formulae describing nonlinear transmission spectra are derived and their relationship to the level of self-similarity in the field profiles is emphasized. In section 5, we propose a design of the nonlinear optical diode based on the differences in bistability thresholds between forward and backward propagation. The efficiency of two schemes is compared: passive, when only one pulse is used, and active, when an additional short pump signal is applied to facilitate switching between stable branches of hysteresis. The last section summarizes the results and presents the conclusions. The appendix provides some details of the FDTD method used in the paper for the dynamical simulation of multilayered structures with instantaneous Kerr nonlinearity.

2. ThM multilayers

There are two main approaches to generate 1D quasi-periodic sequences. The first makes use of a projection from a higher-dimensional space, while the second employs the so-called substitutional sequences [17]. Being strictly quasi-periodic, the Fibonacci sequence can be obtained by using both of the above methods, and the presence of two incommensurable periods

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in their spatial spectra is implicitly related to the projection of a two-dimensional grid onto a one-dimensional line [18]. However, these two approaches are not equivalent to each other, and the use of substitutional sequences tends to be a more general procedure [19]. In particular, this is applicable to ThM sequences and makes it distinct from strictly quasi-periodic sequences. Sometimes it is called a deterministic aperiodic sequence, in order to emphasize that it has more disorder than quasi-periodic ones and stands closer to random sequences [20]. On the other hand, the development of resonances when changing generation number in ThM quasicrystals resembles the analogous development in periodic structures to a certain extent and brings these two kinds of crystal closer to each other [21].

Two dielectric layers of different materials are required to compose ThM quasicrystals. These will be indicated with the letters ‘A’ and ‘B’. They should be arranged in the same way as in literal ThM sequence, which is governed by the following substitutional (‘inflation’) rules: ‘A’ → ‘AB’, ‘B’ → ‘BA’. Thus, starting from a single layer $S_0 = ‘A’$, which is defined to be the ThM quasi-crystal of the zeroth generation or ThM$_0$, one obtains $S_1 = ‘AB’, S_2 = ‘ABBA’, S_3 = ‘ABBABAAB’$ and so forth, with each step giving a sequence of generation number increased by one (figure 1(a)). It can be shown that an additional recurrence relation follows from this definition, which holds for ThM sequences as single blocks: $S_{n+1} = S_n \tilde{S}_n$, where $n$ is the generation number. In this notation $\tilde{S}_n$ indicates a sequence ‘conjugated’ to $S_n$, where all letters are interchanged with their opposite as in the rule ‘A’ ↔ ‘B’.

It is interesting to note that there is only a slight difference between the ThM and Bragg (or periodic) sequences as far as inflation rules are concerned. More specifically, Bragg sequences
possess the inflation rules ‘A’ → ‘AB’ and ‘B’ → ‘AB’. Therefore, the growth rate of these sequences is the same, and the total number of layers is subject to a rapid (exponential) increase as $2^n$. The occurrence of layers ‘A’ and ‘B’ is also the same for both ThM and Bragg structures for any generation number. Nevertheless, such a small modification of the inflation rules leads to completely distinct transmission and localization properties.

The distribution of electric field at resonant frequencies is strictly related to the properties of the specific substitutional sequence that is used to generate a quasicrystal. Figure 1(b) provides an example of how the self-similarity of ThM sequence and its asymmetry for odd generation numbers can manifest themselves in the field profiles. The latter property leads in turn to non-reciprocal behavior of such structures in the nonlinear case. Therefore, we can determine how suitable for maximizing the above effect a particular sequence is by only looking at its literal representation. It is instructive in this regard to compare the ThM sequence with other sequences. The Bragg sequence can always be done mirror symmetric just by dropping the last letter. Taking the third generation as an example, it can be schematically written as $\text{Br}_3 = \text{ABAABABA}|B$. The Fibonacci sequence is also not suitable for the achievement of non-reciprocal behavior, since it is very close to a symmetric sequence [22]. Namely, if the two last letters are dropped, the sequence turns precisely into a mirror symmetric one. For example, the fifth generation can be schematically written as $\text{Fb}_5 = \text{ABA|BA}$. The primary method for computing transmission spectra and field profiles for multilayered structures is the transfer matrix method [23]. If normal incidence is assumed, the electric $E$ and magnetic $H$ fields on the opposite sides of each single layer can be related in the following way:

$$
\begin{pmatrix}
E \\
iH
\end{pmatrix}_{m-1} = M_m \begin{pmatrix}
E \\
iH
\end{pmatrix}_m, \quad (1)
$$

where $d_m$ is the thickness of the layer with refractive index $n_m$ and $\Omega = \omega/c$ is the wavenumber in vacuum. In the representation of the transfer matrix method used in (1) and (2), all imaginary units were transferred to the basis entering it as a factor in front of magnetic field. This allows us to characterize any non-absorbing layered structures by transfer matrices with real elements. In general, the refractive indices of the materials can depend on frequency, but for the sake of simplicity we will assume that all materials are dispersionless. By multiplying together all transfer matrices of subsequent layers, it is possible to construct the total transfer matrix of the structure

$$
M = M_1 M_2 \ldots M_{L-1} M_L \quad (3)
$$

and to find the transmission spectrum in terms of its matrix elements

$$
T(\Omega) = \frac{4}{|M_{11} + M_{22} + i(M_{21} - M_{12})|^2}. \quad (4)
$$

Another—more efficient—approach to obtain transfer matrices of ThM quasicrystals is to make use of the following two-step recurrence relations:

$$
M_{[n+1]} = M_{[n]} \tilde{M}_{[n]}, \quad (5)
$$

$$
\tilde{M}_{[n+1]} = \tilde{M}_{[n]} M_{[n]}, \quad (6)
$$

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supported by the initial conditions $M_{[0]} = M_A$ and $\tilde{M}_{[0]} = M_B$, which are the transfer matrices for layers ‘A’ and ‘B’, respectively. The subscripts in curly brackets indicate generation numbers of corresponding ThM quasicrystals. Formulae (5) and (6) originate from the recurrence relations of the ThM sequence explained above, and stress explicitly the interplay between ‘ordinary’ $S_n$ and ‘conjugated’ $\tilde{S}_n$ counterparts of the sequence. The self-similarity of ThM quasicrystals is naturally embedded in this approach, and this simplifies considerably the computation of transmission spectra for large generation numbers.

However, even this modified representation of the transfer matrix method still contains redundant information about the structure, if one is interested only in the analysis of the properties of the resonance frequencies. In this case, it is sufficient to operate with traces of transfer matrices. The trace is defined as

$$x = \text{Tr}(M) = M_{11} + M_{22}. \quad (7)$$

For ThM quasicrystals of fixed generation numbers, these traces are unchanged under conjugation

$$x_n = \text{Tr}(M_{[n]}) = \text{Tr}(M_{[n-1]}\tilde{M}_{[n-1]}) = \text{Tr}(\tilde{M}_{[n-1]}M_{[n-1]}) = \text{Tr}(\tilde{M}_{[n]}), \quad (8)$$

and for adjacent generation numbers, an additional relation or ‘trace map’ holds [24]:

$$x_{n+2} = \text{Tr}(M_{[n]}\tilde{M}_{[n]}\tilde{M}_{[n]}M_{[n]})$$

$$= \text{Tr}(M^2_{[n]}\tilde{M}^2_{[n]})$$

$$= x_n^2 (x_{n+1} - 2) + 2. \quad (9)$$

Formulae (8) and (9) are valid for $n \geq 1$, and in the derivation of (9) the Cayley–Hamilton theorem was applied to prove that any unimodular matrix satisfies the relation

$$U^2 = x_U U - I, \quad (10)$$

where $x_U$ is the trace of a unimodular matrix $U$, and $I$ is the identity matrix. To use the equation for the trace map (7), two initial conditions are required. They can be found directly from the multiplication of transfer matrices having the form given by equation (2):

$$x_1 = 2 \cos \alpha \cos \beta - (n_A/n_B + n_B/n_A) \sin \alpha \sin \beta, \quad (11)$$

$$x_2 = 2 \cos 2 \alpha \cos 2 \beta - (n_A/n_B + n_B/n_A) \sin 2 \alpha \sin 2 \beta, \quad (12)$$

where $\alpha = n_A d_A \Omega$ and $\beta = n_B d_B \Omega$.

Several important conclusions can be drawn from the trace map of ThM quasi-crystals. The analysis simplifies greatly in the absence of absorption, so that the imaginary parts of the refractive indices are negligible. In this case, all traces will be real, and there will be such spectrum frequencies for which $x_n = 0$. Independent of the value of $x_{n+1}$, this makes $x_{n+2} = 2$, which coincides with the trace of the identity matrix. This simply means that frequencies for which $x_n = 0$ correspond to frequencies of perfect transmission. To prove this, an auxiliary matrix should be constructed, the trace of which gives the difference of diagonal elements of the original transfer matrix:

$$z = \text{Tr}(\sigma_z M) = M_{11} - M_{22}, \quad (13)$$

where $\sigma_z$ is the third (diagonal) Pauli matrix. Provided that $x_n = 0$, it can be readily shown that the diagonal elements of $M_{[n+2]}$ are equal:

$$z_{n+2} = \text{Tr}(\sigma_z M_{[n]}\tilde{M}_{[n]}\tilde{M}_{[n]}M_{[n]}) = \text{Tr}(\sigma_z) = 0. \quad (14)$$
Therefore, the resonances of complete transmission in ThM quasicrystals can be characterized by identity transfer matrices and they are preserved from one generation to another. Only additional resonances can appear in new generations, and the overall transmission spectrum of these quasicrystals shows a fractal nature. As the generation number increases, the spectrum reveals self-similarity and characteristic trifurcation of resonances [24].

In what follows, we will consider that layers ‘A’ and ‘B’ have the same optical thickness, $n_A d_A = n_B d_B$, and we will introduce the reference wavenumber $\Omega_0$, defined as $n_A d_A \Omega_0 = \pi/2$, which corresponds to the quarter wavelength condition. The ratio $\Omega/\Omega_0$ will be called ‘normalized frequency’ and will make the comparison of different structures easier. For example, a normalized frequency $\Omega/\Omega_0 = 1$ corresponds to the middle of the bandgap region for Bragg crystals, but it is the center of a wide ‘pseudo-pass band’ for ThM quasicrystals, a region made of densely located shallow resonances, not always of perfect transmission (figure 2). This band exists only for the specific case of equal optical thicknesses of the layers, and it starts to appear from the second generation, for which formulae (8) and (9) are not valid. The above condition makes them applicable for $n \geq 0$. Moreover, as long as the condition is fulfilled, the transmission spectra of all 1D structures will be translationally invariant along the frequency axis, $T(\Omega/\Omega_0 + 2) = T(\Omega/\Omega_0)$, and symmetric with respect to $\Omega/\Omega_0 = 1$.

3. Bistability and multistability in ThM multilayers

The bistability and multistability properties of Bragg gratings have been studied in depth by a number of authors [25–27]. The knowledge of the field profiles is crucial for the proper
understanding of bistability and multistability in ThM quasicrystals. In correspondence to the
resonance frequencies, these field profiles show a self-similar pattern that strongly resembles
that of a ThM sequence of smaller generation number (figure 1(b)). Due to this fact, these
resonant states are also called lattice-like states [28]. The level of self-similarity depends on
the generation number in correspondence to which a particular resonance first appears. For
example, the field profile shown in figure 1(b) belongs to a ThM quasicrystal of the seventh
generation. It corresponds to the resonance with normalized frequency \( \Omega / \Omega_0 = 0.756 \) and,
according to the bifurcation diagram shown in figure 2, this resonance is inherited from the
fourth generation. Therefore, the field profile mentioned above has the level of similarity equal
to \( 7 - 4 = 3 \), and it is possible to distinguish \( 2^3 = 8 \) independent blocks. Moreover, these blocks
are arranged according to the ThM sequence of the third generation. We have found that for any
ThM structure with layers of equal optical thickness, the level of self-similarity is maximal for
normalized frequencies belonging to the following series:
\[
\Omega / \Omega_0 = \{ 0, 1 - \sigma, 1, 1 + \sigma, 2, 3 - \sigma, 3, 3 + \sigma, \ldots \},
\]
where
\[
\sigma = 1 - \frac{2}{\pi} \arctan \left( \frac{2}{(n_A/n_B) + (n_B/n_A)} \right) \approx 0.512.
\]
However, the localization strength is relatively small at these frequencies, and thus the nonlinear
response cannot be enhanced significantly because the electric field profile is almost flat along
the structure. For this purpose, resonances located near the edges of pseudo-bandgaps are
more suitable. In general, the localization strength is inversely proportional to the level of self-
similarity, and when the length of the structure increases, the resonant states gradually change
from localized to extended ones [29].

The case of Kerr (or cubic) nonlinearity was considered in this work, so that a nonlinear
refractive index was additionally taken into account for each type of layer. This leads to an
intensity-dependent self-phase modulation, which is able to shift resonance frequencies [26].
The direction of this shift is determined by the sign of the Kerr coefficient. The most interesting
cases evidently occur when the sign of the nonlinearity is such that resonances in the spectrum
of transmission bend towards the bandgap regions (figures 3 and 4). In the following, we always
choose positive nonlinear coefficients for the two materials, \( n_{2A} = 2.5 \times 10^{-5} \text{ cm}^2/\text{MW} \) and
\( n_{2B} = 1.0 \times 10^{-8} \text{ cm}^2/\text{MW} \) (see also figures 3 and 4) [30, 31].

One of the most serious advantages provided by the ThM sequence is that for odd
generation numbers the corresponding photonic structures are intrinsically asymmetric, and
nonlinearity is capable of making transmission sensitive to the propagation direction [16].
This feature is completely absent in nonlinear Bragg structures, where hysteresis curves are
the same regardless of the direction of incidence. Although a similar non-reciprocal behavior
can be achieved in the framework of linear optics, the latter typically requires making use of
magneto-optical media with externally applied static magnetic fields [32] or chiral media such
as cholesteric liquid crystals [33]. Moreover, the polarization state is very important for the
correct operation of these linear devices, while the design suggested here is free from such
complications.

The level of self-similarity in the field profiles is also related to the type of hysteresis that
can be observed near a particular resonance. In the presence of several independent localization
centers inside the structure, the interplay between them gives rise to multistability. It is related
mainly to the fact that ThM structures of higher generation numbers can be decomposed into those of lower ones. Therefore, to single out a particular hysteresis type in its pure form, it is necessary to check that the corresponding resonance does not occur in previous generations.

Two characteristic examples are shown in figures 3 and 4. The first case (figure 3) corresponds to a resonance with no self-similarity in the field profile and clearly demonstrates bistability of transmission, whereas in the second case (figure 4) two independent localization centers can be distinguished. The hystereses are shown in figures 3(b) and 4(b); the nonlinear transmission spectra are given in figures 3(c) and 4(c). A detailed explanation of this behavior is given in the next section. Note that, similar to Bragg structures, the maxima of the electric field can be located mostly in layers of one type, so that in principle it would be sufficient to use only one nonlinear material.

4. The coupled resonator model

4.1. Non-separable resonances

In order to provide a qualitative analytical understanding of the nonlinear behavior of ThM quasicrystals, we apply the coupled resonator model, which has been recognized as a useful tool for describing the dynamical properties of single-mode cavities in 2D photonic crystal waveguides [34–36]. Being a phenomenological theory based on general physical concepts like conservation of energy and time-reversal symmetry [37], it is not restricted to the above 2D

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Figure 4. (a) Electric field profiles at resonance frequency $\Omega/\Omega_0 = 0.810$ for the linear ThM$_7$ structure with the same set of parameters as in figure 1(b). (b) Hysteresis of transmission at $\Omega/\Omega_0 = 0.808$. The nonlinear Kerr coefficients used are $n_{2A} = 2.5 \times 10^{-5}$ cm$^2$/MW and $n_{2B} = 1.0 \times 10^{-8}$ cm$^2$/MW. The switching thresholds are different for the forward (blue) and backward (red) incidence. (c) Nonlinear transmission spectra for fixed input intensity $10$ MW/cm$^2$.

In essence, they represent an extension of the scattering matrix method to the time domain. The first equation (17) describes the temporal evolution of a resonant mode in the vicinity of a resonant frequency $\omega_0$. The amplitude of this mode $A$ is normalized in such a way that $|A|^2$ gives the energy inside the cavity. Energy is carried to the cavity from the two ports or channels, which are denoted as $U$ and $V$, located on the left and right sides, respectively (figure 5(a)). The amplitudes of the incident signals are denoted as $u_+$ and $v_+$. They are normalized in such a way that $|u_+|^2$ and $|v_+|^2$ give the power flow. The interaction between these signals and the cavity is described by the two coupling coefficients $\kappa_u$ and $\kappa_v$, respectively. Since the cavity not only accumulates the incident energy but may also partly absorb it or return it back to the channels, there is a damping constant $\gamma$, which characterizes the efficiency of these processes. It can be related to the quality factor of the resonator $Q = \omega_0/(2\gamma)$. As to the influence of...
nonlinearity, it results in the shift of the resonant frequency. In equation (17), we introduce a characteristic energy $P_0$ that allows us to take this shift into account [38], which can be either positive or negative depending on the common sign of the Kerr coefficients in the multilayered structure.

The second equation (18) describes the output from the cavity. The right-hand side of this equation consists of two parts. The first part characterizes the background reflection from the cavity as if the amplitude of the mode inside it were zero. In fact, it has exactly the form of a scattering matrix. The background transmission coefficient $t$ is equal for both directions even for non-symmetric structures, while the reflection coefficients in general have different phases and are denoted as $r_u$ and $r_v$. As to the second part of equation (18), it is proportional to the amplitude of the mode and thus provides the corrections caused by the interaction of incident signals with the cavity.

Assuming that the system operates at a fixed frequency, it is possible to derive from equations (17) and (18) an explicit expression for the nonlinear transmission spectrum along the forward (backward) propagation direction:

$$T^{u,v} \equiv \frac{I_{\text{out}}^{u,v}}{I_{\text{in}}^{u,v}} = \frac{\eta}{1 + (\delta - I_{\text{out}}^{u,v}/I_0^{u,v})^2},$$

where $I_{\text{in}}^u = |u_+|^2$ ($I_{\text{in}}^v = |v_+|^2$) is the input intensity, $I_{\text{out}}^u = |v_+|^2$ ($I_{\text{out}}^v = |u_+|^2$) is the output intensity, $\delta = (\omega - \omega_0)/\gamma$ is the normalized detuning from the resonance frequency, and $\eta = |\kappa_u \kappa_v|^2/\gamma^2$ corresponds to the maximum allowed transmission. The characteristic intensities $I_0^{u,v}$, and this is a crucial observation for the present work, are in general different for the incidence from the left and from the right

$$I_0^{u,v} = (\gamma/\omega_0) P_0 |\kappa_{u,v}|^2.$$

**Figure 5.** (a) The scheme describing the quantities used in equations (17) and (18) for the coupled resonator model. (b) The typical form of the nonlinear transmission spectrum following from formula (19).
In the case of a single Dirac-delta-like nonlinear layer in the system, formula (19) coincides with a previously investigated exact analytical solution [39]. However, taking into account a continuously distributed nonlinearity gives rise to a new feature: if the structure is spatially asymmetric, the characteristic intensity can depend on the direction of incidence, as is evident from equation (20).

Since formula (19) is equivalent to a third-order polynomial in the output intensity \( I_{\text{out}} \), by definition it fails to describe resonances with multistability, but it gives a good approximation for resonances with a strictly bistable response (compare, for instance, figure 5(b) with figures 3(b) and (c).

4.2. Separable resonances

In the more complex case, when the field profiles consist of several independent parts, one simply needs to increase the number of equations in the coupled resonator model. There should be exactly one set of equations (17) and (18) for each separable resonance. For example, the case shown in figure 4 can be described by the following system of equations:

\[
\begin{align*}
\frac{dA}{dt} &= -\left[ i\omega_0 \left( 1 + \frac{|A|^2}{P_a} \right) + \gamma_a \right] A + \kappa_a (u_+ + s_+), \\
\frac{dB}{dt} &= -\left[ i\omega_0 \left( 1 - \frac{|B|^2}{P_b} \right) + \gamma_b \right] B + \kappa_b (v_+ + s_-), \\
(u_-) &= \exp(i\varphi_a) \begin{pmatrix} u_+ \ \\ s_+ \end{pmatrix} + A\kappa_a, \\
(v_-) &= \exp(i\varphi_b) \begin{pmatrix} v_+ \ \\ s_- \end{pmatrix} + B\kappa_b,
\end{align*}
\]

(21)–(24)

where \( A \) and \( B \) give the amplitudes of the two separable resonances, which interact through a virtual port \( S \) (figure 6(a)). These resonances have the same resonance frequency \( \omega_0 \), but in general their damping constants \( \gamma_a, \gamma_b \) and characteristic energies \( P_a, P_b \) will be different. Since both separable resonances shown in figure 4 are symmetric, the coupling to either of them can be described by a single coefficient \( \kappa_a, \kappa_b = \sqrt{\gamma_a, \gamma_b} \). Moreover, because the common resonant frequency is located in the bandgap (or pseudo-bandgap) region, the background transmission can be neglected, and the scattering matrix entering equation (18) can be replaced by a reflection coefficient, which is equal for both directions of incidence and has the form \( \exp(i\varphi_{a,b}) \).

The system (21)–(24) can be readily solved in the frequency domain. We found the following formula, which describes the nonlinear transmission spectra with multistable behavior:

\[
T = \frac{1}{|\xi_a\xi_b + (1-i\xi_a)(1-i\xi_b)\exp[-i(\varphi_a + \varphi_b)]|^2}.
\]

(25)

Expression (25) contains two parameters, \( \xi_a \) and \( \xi_b \), which can be calculated given the output signal \( v_- \) (or \( u_- \)) by using the following ladder:

\[
B = v_- / \kappa_b.
\]

(26)
Figure 6. (a) The scheme describing the quantities used in equations (21)–(24) of the coupled resonator model. (b, c) The typical form of nonlinear transmission spectra following from formula (25), when the dimensionless parameters are set to $\omega_0 = 800$, $\gamma_a = 2.89$, $\gamma_b = 1.44$, $P_a = 120$ and $P_b = 80$. The only difference between (b) and (c) is in the phase mismatch $\varphi_a + \varphi_b = 0.05\pi$ and $\varphi_a + \varphi_b = \pi$, respectively. The amplitudes of incident signals are set as $u_+ = 2$ for the propagation in the forward direction (blue lines) and $v_+ = 2$ for the backward direction (red lines). The linear transmission spectra are given by gray lines.

\[ \xi_b = \delta_b + \frac{\omega_0 |B|^2}{\gamma_b P_b}, \]  
\[ A = (1 - i\xi_b + i\xi_b\exp[i(\varphi_a + \varphi_b)])(v_- / \kappa_a), \]  
\[ \xi_a = \delta_a + \frac{\omega_0 |A|^2}{\gamma_a P_a}. \]  

The typical nonlinear transmission spectra calculated by using formula (25) are shown in figures 6(b) and (c). The main parameter that determines the shape of nonlinear transmission spectra is the phase mismatch $\varphi_a + \varphi_b$. It describes the efficiency of interaction between separable resonances and is responsible for the splitting of initially identical resonant frequencies even in the linear case. The only difference between figures 6(b) and (c) is in the phase mismatch, and the two well-separated resonances shown in the first case become indistinguishable for small intensities in the second case (see gray lines). As the intensity of the incident signal increases, this degeneracy disappears, giving rise to multistability (see red and blue lines for the forward and backward incidence, respectively). Qualitatively, the nonlinear transmission spectra shown in figure 6(c) correspond to that in figure 4(c).

It is worth noting that if $\varphi_a + \varphi_b = 0$, formula (25) reduces to

\[ T = \frac{1}{1 + (\xi_a + \xi_b)^2}, \]  

which means that the two resonances do not interact with each other and the resulting nonlinear transmission spectra reveal only a bistable behavior.
5. Nonlinear optical diode action in ThM multilayers

The difference in bistability thresholds can be small, but it creates favorable conditions for unidirectional propagation, so that ThM quasicrystals can be used as all-optical diodes [16]. Similar to electronic circuits, these devices are indispensable when there is a need to suppress the flow of light in one direction or to avoid problems caused by unwanted reflections. Various types of optical diodes have been proposed and realized, which can work both in the linear [32], [33] and in the nonlinear regime [40–43]. The primary figure of merit, which determines the efficiency of this device, is the contrast ratio $C = T_f/T_b$ between transmission along the forward ($T_f$) and backward ($T_b$) directions. $C$ can be very large in ThM structures due to the fact that this device can operate on two different hysteresis branches depending on propagation direction (figure 3(b)).

It is possible to use the intensities of both up and down transitions to achieve strongly non-reciprocal transmission. In the former case, the scheme works in the passive mode, but the maximum value of transmission is limited [44]. In the latter case, the transmission can be almost perfect, but the scheme requires an additional short pump signal in order to switch to the higher stable branch of hysteresis [45].

The FDTD method was applied to demonstrate the switching dynamics in the time domain (figure 7). To maintain the second-order accuracy of the Yee scheme in the case of multilayered structures, a non-uniform spatial mesh was used with electric field nodes aligned to the boundaries between layers (see the appendix). Since the structure can accumulate and release
energy, the sum of reflected (given by red dotted line in figure 7) and transmitted (blue dashed line) intensities should not necessarily give the incident one (gray solid line). This constraint becomes valid only after all the transient dynamics is over.

The input intensity was set first to the value, which is sufficiently large for the upswitching in the forward direction, but at the same time is smaller than the corresponding value for the backward direction. The maximum transmission obtained was $T_f = 65\%$ with the contrast ratio $C = 7.2$, which is quite close to the theoretical limit of $C_{\text{max}} = 9$ following from (31) in the assumption of the passive scheme [44]. In the second stage, the input intensity was set just above the down switching threshold for the forward direction, which ensures transmission of almost $T_f = 100\%$ in this direction, while in the opposite direction transmission will be strongly suppressed due to the presence of the pseudo-bandgap. The contrast ratio achieved was $C = 20$, but it is not limited in principle. The major drawback of this scheme is that it is necessary to force the system to switch to the upper branch of hysteresis whether by applying an auxiliary pump signal or by temporarily increasing the input intensity.

Since highly nonlinear materials such as polymers can be easily damaged by strong input pulses, it is important to check whether the parameters used are practical. The simplest approach to do that is to estimate the nonlinear changes of the refractive index inside the structure. If they are small, the sample will not be damaged, and other effects such as saturation of nonlinearity can be neglected. For this purpose, we recall figure 3(a), which gives the electric field profile at resonance used in the simulation, and determine that the local enhancements of the field inside the structure are about $f = 7$ times in comparison with the incident signal. As an averaged value of input intensity, we choose $I = 10\, \text{MW/cm}^2$, although this value is not specific and can be lowered significantly together with the switching thresholds by changing the detuning from resonant frequency. Taking into account that the largest nonlinearity in the system is associated with the layers of polydiacetylene 9-BCMU with $n_{2A} = 2.5 \times 10^{-5} \, \text{cm}^2/\text{MW}$, we can estimate that the nonlinear changes of the refractive index are of the following magnitude: $\Delta n = n_{2A} \times f^2 \times I \approx 10^{-2}$. Therefore, we expect that the structure will be stable.

6. Conclusions

The nonlinear properties of quasicrystals based on ThM sequence were investigated. It was shown that the interplay between the intrinsic spatial asymmetry of these structures for odd generation numbers and Kerr nonlinearity makes the switching thresholds induced by bistability and multistability sensitive to the propagation direction. The role of self-similarity was emphasized to explain the shape of hysteresis curves observed near a particular resonance, and conditions necessary to achieve highly non-reciprocal propagation were formulated. The coupled resonator model was used as a phenomenological model. FDTD simulations have been used to confirm the results obtained by the nonlinear transfer matrix method, and to show how ThM multilayers can exhibit an effective optical diode action, both in passive and active operation.

Acknowledgment

This work was supported by the German Max Planck Society for the Advancement of Science (MPG).

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Appendix

Maxwell’s equations in differential form
\[
\frac{\partial E_z(x, t)}{\partial x} = \frac{1}{c} \frac{\partial H_y(x, t)}{\partial t},
\]
\[
\frac{\partial H_y(x, t)}{\partial x} = \frac{1}{c} \frac{\partial D_z(x, t)}{\partial t},
\]
(A.1)
(A.2)

can be approximated by finite differences
\[
H_y^{n+1/2}(i + 1/2) = H_y^{n-1/2}(i + 1/2) + \frac{c \Delta t}{\Delta x} \left[ E_z^n(i + 1) - E_z^n(i) \right],
\]
(A.3)
\[
D_z^{n+1}(i) = D_z^n(i) + \frac{c \Delta t}{\Delta x} \left[ H_y^{n+1/2}(i + 1/2) - H_y^{n+1/2}(i - 1/2) \right],
\]
(A.4)

where integer indices \( n \) and \( i \) serve as temporal and spatial coordinates for the Yee scheme \([46]\).

The Courant stability condition requires \( c \Delta t \leq \Delta x \), and it will be assumed that \( c \Delta t = \Delta x/2 \).

To tailor two different uniform grids, it is convenient to use the integral representation of Maxwell’s equations
\[
E_z(x_R, t) - E_z(x_L, t) = \frac{1}{c} \frac{\partial}{\partial t} \int_{x_L}^{x_R} H_y(x, t) \, dx,
\]
(A.5)
\[
H_y(x_R, t) - H_y(x_L, t) = \frac{1}{c} \frac{\partial}{\partial t} \int_{x_L}^{x_R} D_z(x, t) \, dx.
\]
(A.6)

After integration over the boundary between two layers, the following finite difference equations can be obtained (the symbols used are defined in figure A.1).
\[
H_y^{n+1/2}(\beta_b) = H_y^{n-1/2}(\beta_b) + \frac{c \Delta t}{\rho_a + \rho_b} \left[ E_z^n(\rho_b) - E_z^n(-\rho_a) \right],
\]
(A.7)
\[
E_z^{n+1}(-\rho_a) = E_z^n(-\rho_a) + \frac{c \Delta t}{\varepsilon_a \rho_a + \varepsilon_b \beta_b} \left[ H_y^{n+1/2}(\beta_b) - H_y^{n+1/2}(-\beta_b) \right].
\]
(A.8)

It can be shown that these equations are valid up to the second order if the nodes of the electric field are placed on the boundaries between layers, and the steps of the spatial grids inside layers of different types satisfy \( \varepsilon_a (\Delta x_a)^2 = \varepsilon_b (\Delta x_b)^2 = (\Delta x)^2 \). This condition can be exactly fulfilled if layers composing the structure are of equal optical thickness. In the general case, the second-order error of the scheme with a non-uniform mesh is estimated to be of the order of
\[
\varepsilon_b (\Delta x_b/m_b) \delta,
\]
(A.9)

where \( m \) is the number of nodes inside an unmatched layer and \( \delta \) is a missing or superfluous thickness that caused the mismatch. When the scheme with uniform mesh is used, the error is proportional to the difference in permittivities and cannot vanish
\[
(\varepsilon_b - \varepsilon_a) (\Delta x/2)^2.
\]
(A.10)
The scheme describing the quantities used in the tailoring of two uniform grids at the boundary between dielectric media.

The constituent relation in the presence of instantaneous Kerr nonlinearity is considered

\[ D_z = \varepsilon_L E_z + \varepsilon_K E_z^3. \]  

(A.11)

According to (A.6) and (A.8), the linear \( \varepsilon_L \) and nonlinear \( \varepsilon_K \) contributions to permittivity should be averaged at the boundaries between layers

\[ \varepsilon_L,_{\text{eff}}(i) = (\varepsilon_{L,a} \Delta x_a + \varepsilon_{L,b} \Delta x_b) / \Delta x, \]  

(A.12)

\[ \varepsilon_K,_{\text{eff}}(i) = (\varepsilon_{K,a} \Delta x_a + \varepsilon_{K,b} \Delta x_b) / \Delta x. \]  

(A.13)

Equation (A.11) can be written in finite differences as

\[ E_n = [D_n + 2\varepsilon_K (E_{n-1})^3] / [\varepsilon_L + 3\varepsilon_K (E_{n-1})^2], \]  

(A.14)

which is equivalent to solving the cubic equation (A.11) by iteration and is always stable for positive \( \varepsilon_L \) and \( \varepsilon_K \). In contrast to the equation

\[ D_z = \varepsilon_L E_z + (3/4)\varepsilon_K |E_z|^2 E_z, \]  

(A.15)

the use of constituent relation (A.11) allows taking into account the generation of third harmonic, but attention should be paid to the correct definition of \( \varepsilon_K \).

To separate incident and reflected waves from the structure, the so-called total-field scattered-field technique is used on the left (and of the right) of the structure

\[ H_y^{n+1/2}(i_F \mp 1/2) = H_y^{n-1/2}(i_F \mp 1/2) \mp \frac{c \Delta t}{\Delta x} \left[ E_z^n(i_F) - E_z^n(i_F) - E_z^n(i_F) \mp 1 \right], \]  

(A.16)

\[ D_z^{n+1}(i_F) = D_z^{n}(i_F) + \frac{c \Delta t}{\Delta x} [H_y^{n+1/2}(i_F + 1/2) - H_y^{n+1/2}(i_F - 1/2) \mp H_y^{n+1/2}(i_F \mp 1/2)]. \]  

(A.17)

The sources of the electromagnetic field on the left (and on the right) are introduced as

\[ E_z,_{\text{inc}}(i_S) = E_z,_{\text{inc}}(nc \Delta t), \]  

(A.18)

\[ H_y,_{\text{inc}}(i_S \mp 1/2) = \mp E_z,_{\text{inc}}((n + 3/2)c \Delta t). \]  

(A.19)
The absorbing boundary conditions in the case when $c \Delta t = \Delta x / 2$ take the following form on the left (and on the right) boundary of the simulation region:

$$H_y^{n+1/2}(i_B \mp 1/2) = H_y^{n-3/2}(i_B \pm 1/2). \quad (A.20)$$

Smooth envelopes for switching on and off monochromatic signals are defined by splines of sixth order, which ensure the continuity of fields up to the second derivative [48]

$$U(t) = \begin{cases} 
  u(t/\tau), & \text{for } 0 < t < \tau, \\
  1, & \text{for } \tau \leq t \leq T - \tau, \\
  u[(T - t)/\tau], & \text{for } T - \tau < t < T,
\end{cases} \quad (A.21)$$

where $T$ is the total time when the signal is nonzero, $\tau$ is the duration of the switching and $u(x) = 10x^3 - 15x^4 + 6x^5$. The intensities of reflected and transmitted waves $A$ can be extracted from the simulation through the following integration:

$$(1/P) \int_{t}^{t+P} A \cos(m\omega t - \varphi) \exp(in\omega t) \, dt = (A/2) \exp(i\varphi) \delta_{nm}, \quad (A.22)$$

where $P = 2\pi/\omega$ is the period of the carrier wave.

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