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Two-Wave Mixing in Organic-Inorganic Hybrid Structures for Dynamic Holography

Vera Marinova, Shiuan Huei Lin and Ken Yuh Hsu

Abstract

The chapter reviews recent progress of two-wave mixing in a novel organic-inorganic hybrid structures that combine essential properties as large anisotropy and strong birefringence, typical for organics with the excellent photosensitivity and photoconductivity of inorganics into single, compact devices. Depending on the designed assembly and operation principle, the proposed structures can record dynamic holographic gratings at Raman-Nath or Bragg regimes of diffraction, respectively. When the two beams interact in a structure based on a photoconductive material and birefringent layer (usually liquid crystal), the beam coupling with high amplification values occur in a liquid crystal layer; however, the fringe period of recorded holograms is limited to few μm scale. In contrast, when the two beams interact in a structure based on a photorefractive material and birefringent layer, the beam-coupling occurs in both composites, due to the surface activated photorefractive effect. The prime significance of the later structure is the ability to act as a holographic grating at Bragg regime allowing sub-micron spatial resolution. Moreover they are easy and simple to fabricate where the processes are all optically controlled. The above examples open scenarios to design new devices that meet the latest requirements of 3D display technologies and optical information processing.

Keywords: liquid crystals, inorganic crystals, beam amplification, Raman-Nath diffraction, Bragg diffraction, photorefractive effect, space-charge field

1. Introduction

The demand to develop advanced functional devices with fast operation speed, high memory capability, submicron spatial resolution and low energy consumption continuously tends to increase. For example, to reach a wide viewing angle, the pixel pitch of the recording media
needs to be comparable or smaller than the wavelength of the light. Another approach is to
design all optically controlled devices for light control and manipulation. All these features
make the combination between outstanding properties as large anisotropy and strong birefrin-\ngence typical for organics (offering easy processing, large structural flexibility and low cost)
with the excellent photosensitivity and photoconductivity of inorganic materials (providing
mechanical stability and energy gap manipulation) attractive to design single, compact struc-
tures with enhanced functionality. Moreover, an emergent need of devices sensitive at the
near-infrared spectral range (the required illumination for biological and medical samples) is
of particular importance for biomedical sensing and with significant impact for the society.
In that aspect, the holography, due to its unique nature, is expected to play an essential role.

The following chapter is focused on the two-wave mixing in organic-inorganic hybrid struc-
tures and dynamic holography recording resulting to formation of light-induced gratings.
The two-wave mixing (denoted also as a two-beam coupling) takes place in a variety of non-
linear media as photorefractive materials (the photorefractive effect refers to refractive index
modulation in response to light), third-order non-linear media (as Kerr media) or semicon-
ductor amplifiers. The two-wave mixing is expressed by two beam interactions, forming an
interference pattern, which is characterized by periodic spatial variation of the light-intensity
distribution. Its main significance is the ability of unidirectional optical energy exchange
between the two beams (gain amplification) allowing weak beam to grow exponentially with
the distance, which opens various opportunities for designing a new structures and elements
for practical realizations.

Depending of the organic-inorganic hybrid structure design and in more particular how the
inorganic material control the dynamic grating formation, they can operate at Raman-Nath or
Bragg match regime of diffraction:

i) Generally, the organic-inorganic structures are assembled by photoconductive material
(usually inorganic crystal) and birefringent material (liquid crystal [LC] or polymer
dispersed liquid crystal [PDLC]). Their operation principle relays on electro-optically
controlled birefringence of the liquid crystal molecules that allows spatial modulation of
the amplitude or the phase of incident beam. In such configuration, the inorganic crystal
serves as a photoconductive layer, which controls the LC molecules alignment and allows
subsequent light modulation. As a result, the two-wave mixing happens in a LC layer
with very high amplification values; however, the fringe period of the recorded holo-
grams is limited to few micrometer scales (Raman-Nath regime of diffraction). Usually,
this type of hybrid structures is known as electro-optically controlled devices.

ii) Recently, a novel type of organic-inorganic structures has been proposed assembled by
photorefractive material and birefringent layer (LC or PDLC). Their operation principle
relays on surface-activated photorefractive effect and more specific on the photo-generated
space charge field, acting as a driving force for LC molecules reorientation and subse-
dually the refractive index modulation. The prime significance is the fact that the two-
wave mixing happens in both photorefractive and birefringent layers, where the charge
carrier migration, high-trap density and space-charge field come from the inorganic substrate, whereas the beam amplification is provided by the LC layer. As a result, the proposed hybrid structures act as dynamic holographic grating at Bragg match regime allowing sub-micron spatial resolution. Moreover, the above configuration is easy and simple to fabricate (no need of conductive layer deposition [and in a case of PDLC, no need of alignment layers and polarizers]), and all the processes are controlled only by the action of light. Therefore, they are noted as all optically controlled hybrid devices.

The chapter is organized as following: at the beginning, a brief introduction of two-wave mixing and gain amplification phenomena as a consequence from the non-linearity of photorefractive effect will be reviewed. The main significances of the optical energy exchange and phase shift between the interference pattern and refractive index grating are discussed. Next, the two-wave mixing in two types of organic-inorganic hybrid structures (electro-optically controlled and all optically controlled) will be presented and compared. Examples as optically addressed spatial light modulation devices; in dynamic holography and image processing are demonstrated and discussed. In concluding remarks, further prospective to design varieties of novel all optically controlled hybrid devices will be discussed.

2. Raman-Nath and Bragg match regimes of diffraction

The interaction between two coherent laser beams inside the photosensitive material generates a light-intensity fringe pattern of bright and dark regions (sinusoidal light intensity pattern) expressed by [1, 2]:

$$I(x) = I_0 [1 + m \cos(Kx)]$$ (1)

where $I_1$ and $I_2$ are the intensities of both beams, $I_0$ is the total intensity $I_0 = (I_1 + I_2)$ and $m$ is the light modulation, $m = 2 \frac{I_1 I_2}{I_0}$.

The created diffraction grating is defined by the two interfering beams and their spatial coordinates, determined by the grating wave vector $K = 2 \pi / \Lambda$ and the spatial fringe period $\Lambda = \frac{\lambda}{2 \sin \theta}$, where $\lambda$ is the wavelength, and $\theta$ is the external half angle between of the intersection beams. After propagation through the medium, the same beams diffract from the holographic grating, which they formed.

The light diffraction phenomenon from periodic structures has been extensively discussed in the past and defined to two regimes of diffraction: (i) Raman-Nath regime (when several diffracted waves are produced named thin grating) and (ii) Bragg matched regime (when only one diffracted wave is produced named thick or volume grating) [1, 2].

Briefly, in Raman-Nath regime, after interacting with the grating, the incident beam is split into several beams resulting in different orders of diffraction (see Figure 1(a)). As a consequence, there are several diffracted waves produced $0, \pm 1, \pm 2, \pm 3, \ldots \pm m$, which correspond
to the wave vectors $k$, $k \pm K$, $k \pm 2K$, $k \pm mK$, where $k$ is the wave number of light beam in the medium and $K$ is the grating wave vector. The diffraction efficiency for the Raman-Nath diffraction is given by [1]

$$\eta_m = |J_m(\delta)| = J_m^2 \left( \frac{2\pi n L}{\lambda \cos \theta} \right)$$  \hspace{1cm} (2)

where $J_m(\delta)$ is the amplitude of the $m$th order diffracted beam, expressed by Bessel’s function ($\delta$ is expression of modulation index corresponding to the multiple scattered orders), $n$ is the refractive index of the medium and $L$ is the interaction thickness [3, 4].

In a Bragg regime, after interacting with the grating, only one diffracted wave is produced, and the diffraction occurs only when the incident angle satisfies the Bragg conditions [1–4] (both the energy and momentum are conserved) — see Figure 1(b).

The simplified diffraction efficiency (in case of transmission type, sinusoidal phase grating) is given by [2]

$$\eta = \sin^2 \left( \frac{2\pi n L}{\lambda \cos \theta} \right)$$  \hspace{1cm} (3)

Generally, Klen-Cook dimensionless parameter has been accepted as distinguishing factor between the Raman-Nath or Bragg regimes of operations, defined by [5]:

$$Q = \frac{2\pi n L}{n \Lambda^2}$$  \hspace{1cm} (4)

Although this parameter has been extensively used as a criterion which regime to apply ($Q \leq 1$ for Raman-Nath or $Q > -1$ for the Bragg-matched regime), it requires several limitation of the grating strength [6].
3. Two-wave mixing and optical gain amplification in photorefractive media

The two-wave mixing appears in variety of non-linear media, and owing to its wide range of applications, it has been extensively studied in past [7–10]. In general, the two-wave mixing is described by two beam interactions inside the photosensitive material, forming a light interference pattern (index grating). As a result, the two beams diffracted by the index grating they created in a way that in one direction, the diffracted and transmitted intensities provide constructive interference (with higher resultant intensity), whereas in the other direction, the beams experience destructive interference (with lower resultant intensity). Thus, the most significant importance of the two-wave mixing is the energy exchange between the two interacting beams.

The light illumination in a photosensitive material causes generation of free charge carriers and their redistribution from the regions of high intensity to those of low intensity (see Figure 2). This net migration leads to inhomogeneous charge distribution and accumulation of an internal electric field known as a space charge field \( E_{s} \). Actually, this space charge field is of key importance for the photorefractive effect and can play significant role for the LC molecules reorientation as discussed further.

Briefly, the space charge field is expressed by the following set of well-known equations [1, 7, 8]:

\[
\frac{\partial N}{\partial t} - \frac{\partial N_D}{\partial t} = \frac{1}{e} \nabla \cdot J \tag{5}
\]

\[
\frac{\partial N_D}{\partial t} = (N_D - N_D^t) s I - \gamma_R N N_D^t \tag{6}
\]

\[
J = J_{\text{drift}} + J_{\text{diffusion}} = eN\mu E_{sc} + k_B g \mu N \nabla N \tag{7}
\]

\[
\nabla \cdot \varepsilon E_{sc} = \rho(r) = -q(N + N_A - N_D^t) \tag{8}
\]

where \( e \) is the electron charge, \( N \) is density of main charge carriers, \( N_D \) is total donor density, \( N_D^t \) is ionized donor density, \( N_A \) is density of acceptors, \( s \) is absorption cross section of excitation, \( \gamma_R \) is ionized trap recombination rate, \( \mu \) is mobility, \( J \) is current density, \( \rho \) is charge density, \( \varepsilon \) is dielectric constant, \( k_B \) is Boltzmann constant and \( T \) is the temperature. In Eqs. (5)–(8), Eq. (5) is the rate equation of the main carriers density; Eq. (6) is the rate equation of ionized donors (the first term is the rate of main carriers generation and the second term is the rate of the trap capture); Eq. (7) is the current density equation (if neglect the photovoltaic effect); and Eq. (8) is the Poisson equation.

The first term in Eq. (7) is expressed by the drift of the charge carriers due to the space charge field \( E_{sc} \) and the second term is a diffusion, due to the gradient of the charge carrier density, expressed by the diffusion length \( l_D = \left( \frac{2e^2 \mu T_D}{k_B T} \right)^{\frac{1}{2}} \); where \( T_D = \frac{e^2 N_A}{4\pi^2 \mu k_B T} \) is the diffusion time and
Figure 2. The photorefractive effect: (a) two-beams interference; (b) photo excitation process (intensity pattern); (c) charge transport; (d) space-charge distribution; (e) space-charge field and (f) index grating formation.
\[ E_D = \frac{kD}{p} = \frac{k_s}{E} \] is the diffusion field. The magnitude of the \( E_\text{sc} \) depends on several materials parameters, among which the Debye screening length \( l_D = \sqrt{\frac{\varepsilon \varepsilon_0}{N_{\text{eff}} k_B T e^2}} \) and effective trap density \( N_{\text{eff}} = \frac{(N_i - N_D)N_D}{N_D} \).

In liquid crystals, the drift is the dominant mechanism for the charge carrier migration due to the small trap density of organics. In contrast, the diffusion is the dominant mechanism for inorganic materials and the rate at which the recombination happens determine how far the main charge carrier diffuse and how strong is the refractive index modulation. For instance, inorganic crystals offer several orders of magnitude higher concentration of effective trap density in contrast to the LCs and therefore are able to support formation of the small grating spacing and Bragg match regime of diffraction as will be discussed later.

As a result of charge migration and redistribution, the space charge field in combination with the electro-optic effect modulates the refractive index of the media via the Pockel’s effect [1, 8, 9]:

\[ \Delta \left( \frac{1}{n^2} \right) = r_{ijk} E_{\text{sc}} \] (9)

where \( r_{ijk} \) is the electro-optic coefficient.

The recorded refractive index grating can diffract light, with the diffraction pattern reconstructing the light-intensity pattern, originally stored in the media. Therefore, the index grating created in the photorefractive material is a “volume phase hologram”, which can be written and erased by light, making photorefractive materials fully reversible. Thus, the photorefractive materials have the ability to detect and store spatial distributions of optical intensity in the form of spatial patterns of modulated refractive index.

The most significant consequence from the photorefractive effect is the phase shift between the light-intensity pattern and internal spatial pattern where the later one shifted in respect to the intensity distribution by \( \pi/2 \) period (see Figure 2). This \( \pi/2 \) phase shift induces an optical energy exchange between the two interacting beams (beam amplification) and refers the photorefractive effect as non-local, non-linear effect [2, 7, 8]. Therefore, when two interfering beams have different intensities noted as respectively \( I_s \) “signal” beam (with lower intensity) and \( I_p \) “pump” beam (with higher intensity), due to destructive and constructive interference, the unidirectional transfer of optical energy allows a weak beam to grow exponentially with the distance. As a result, at the exit of the medium, the signal beam is not only amplified but also experienced a non-linear phase shift.

The interaction between the two coherent laser beams inside the photosensitive material (assuming the grating wave vector \( \mathbf{K} \) directed along the x axis) gives the total electric field of the two incident beams

\[ E = A_1 \exp[i(k_1 \cdot r - \omega_1 t)] + A_2 \exp[i(k_2 \cdot r - \omega_2 t)] \] (10)
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voltage of 20 V (for 7.3-μm droplet size) of PDLC layer, the threshold electric field is
\[ E_{th(PDLC)} = 2 \times 10^6 \text{ (V/m)} \]. Substituting the known values into the boundary condition
\[ E_{sc(BSO:Ru)} \times \varepsilon(BSO) = E_{sc(PDLC)} \times \varepsilon(PDLC) \], we found the required electric field to re-orient LC molecules in PDLC
layer is \( E_{sc(PDLC)} = 1.25 \times 10^5 \text{ (V/m)} \). Obviously, the generated \( E_{sc} \) field inside BSO:Ru substrate is
strong enough to penetrate into the PDLC layer and realign the LCs molecules.

For BTO:Rh/LC structure, similar procedure has been performed. The estimated space
charge field in BTO:Rh substrate is about \( E_{sc(BTO:Rh)} = 1.9 \times 10^5 \text{ (V/m)} \). Taking into account the
experimentally measured driving voltage of LC layer as 2 V (for 12-μm thickness) and the
threshold electric field as \( E_{th(LC)} = 1.67 \times 10^5 \text{ (V/m)} \), the required space charge field is about
\( E_{sc(LC)} = 1.5 \times 10^5 \text{ (V/m)} \).

The above two examples verify that the photo-induced space charge field \( E_{sc} \) created in pho-
torefractive substrates can grow high enough to exceed the threshold electric field and pen-
etrate into the birefringent layer. The advantage of using PDLC over the LC is no need of
alignment layer (since the polymer binder defines the droplets orientations), which permits
the photo-generated space-charge field to penetrate directly into the PDLC layer.

Figure 10 shows the experimentally measured gain coefficient \( \Gamma \) (cm\(^{-1}\)) dependence on the
grating spacing \( \Lambda \) (μm) when the ratio of signal-to-pump beam is 1/70. Besides, Figure 10 shows the theoretically simulated strength of the space-charge field \( E_{sc} \) displayed as a single line. For BSO:Ru/PDLC structure, the measured beam amplification value of \( \Gamma = 45 \text{ cm}^{-1} \) is
almost three times higher than reported for similar hybrid structures using double-side pho-
torefractive substrates as CdTe (16 cm\(^{-1}\)) and GaAs (18 cm\(^{-1}\)), operating at near infrared spectral range [46, 47]. In case of BTO:Rh/LC structure, \( \Gamma \) reached almost 10 cm\(^{-1}\) at 1-μm grating spacing. It is assumed that large amplification effect comes from suitable doping elements (as Ru and Rh) addition in sillenite crystal structure, which provides enough density of trap levels to support accumulation of high-resolution space charge field.

![Figure 10](https://example.com/figure10.png)

**Figure 10.** (a) Gain coefficient dependence on the grating spacing \( \Lambda \) (μm) and (b) Theoretical simulation of \( E_{sc} \) dependence on \( \Lambda \) (μm) for BSO:Ru/PDLC structure.
As discussed earlier, the Debye screening length in photorefractive material need to be very small to support high concentration of effective trap density. For BSO:Ru crystal, the Debye screening length of 0.08 μm has been calculated from Eq. (7) and materials parameters in Table 1. Moreover, as shown in Figure 10, the $E_{sc}$ decreases with increasing the grating spacing from 1 to 2 μm. As the gain amplification is controlled by the space charge field, it follows the behaviour of the $E_{sc}$ on the way to decrease with increasing the grating spacing $\Lambda$. In that aspect, multi-layer structure may increase the effective interaction length and optimize the $E_{sc}$ penetration depth (however limited by scattering losses).

5.3. Applications

The prime significance of the reviewed hybrid structure is all optically controlled processes. For example, the switching ability BSO:Ru/PDLC structure is demonstrated at Figure 11. An image pattern (rectangular mask) is placed into the input plane of 4-f optical system, and the structure is illuminated with 1064-nm Gaussian beam. When the pump light illuminates the device, the PDLC layer transparency is changed due to the induced space charge field in the photorefractive substrate. Therefore, by controlling the droplet size and consequently the driving voltage of the LC molecules from one side and optimizing the charge carriers’ concentration in crystal matrix (providing high enough density for high resolution space charge field), the proposed structure can be further optimized. Moreover, the beam coupling can be significantly improved by addition of nanoparticles in LC layer, which affects the dielectric anisotropy and decreases the driving (threshold) voltage.

6. Conclusion

The chapter reviewed recent progress of two-wave mixing and beam amplification in novel type of hybrid structures that combine photoconductive and photorefractive properties of
inorganic crystals together with the high birefringence and anisotropy of LC (or PDLC) layers. When inorganic crystal acts as photoconductive substrate, the photo-generated charge carriers control the LC molecules alignment and subsequent light modulation. As a result, the two-wave mixing happens in a liquid crystal layer (the active layer) with high amplification values; however, the fringe period of the recorded gratings is limited to few micrometers. When inorganic crystal acts as photorefractive substrate, the two-beam coupling happens at both the photorefractive substrate and LC layer where the space charge field is the driving force for LC molecules reorientation and refractive index modulation. In this configuration, all the processes are controlled by light-permitting submicron resolution.

Depending on the designed structure assembly and operation principle, dynamic holographic gratings at Raman-Nath or Bragg match regimes of diffraction can be recorded. The large trap density and small grating spacing typical for photorefractive materials allow to reach the Bragg matched conditions in all optically controlled structure in contract to the large grating spacing and small trap density typical for LC-based electro-optically controlled structures, which support the Raman-Nath regime of diffraction. Furthermore, by selecting the photosensitivity of inorganic substrate, the spectral interval can be easily adjusted in a regions, where the LC molecules (or PDLC) are not enough sensitive.

The proposed organic-inorganic hybrid structures can control transmission, reflection and scattering of light and are considered in playing an essential role in 3D holographic display technologies and providing sub-micron spatial resolution, large viewing angle and low driving voltage. The reviewed examples can be classified as novel type of non-linear optical components, which exhibit attractive capabilities for light manipulation, coherent image amplification, to control the group velocity of modulated signal beam, in video display technologies. The advantage of simple fabrication and compactness open the way to design varieties of new structures and elements that meet the up-to-date requirements of 3D display technologies and optical information processing.

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