

Module 4 : Third order nonlinear optical processes

Lecture 25 : Optical phase conjugation and degenerate four-wave-mixing

Objectives

In this lecture you will learn the following

- Outline of the physical phenomena involved.
- Theory of degenerate four-wave-mixing (DFWM) and Optical phase conjugation.
- Equivalence of DFWM to real time holography.
- Applications in
 - Wavefront recovery,
 - Imaging and
 - Optical computing.

Optical phase conjugation and degenerate four-wave-mixing:

One of the most remarkable and revolutionary experiments in nonlinear optics was undoubtedly the first observation of the phase conjugation and its distortion correction or wave front- restoring properties. This was performed by Zel'dovich et-al [1] in 1972 at the Lebedev Institute. Trying to understand their earlier observation that the back scattered wave in stimulated Brillouin scattering(SBS) propagated in same solid angle as the input wave, they looked at the correlation between input and output wave fronts. The experimental setup used for this is shown schematically in Figure 25.1.

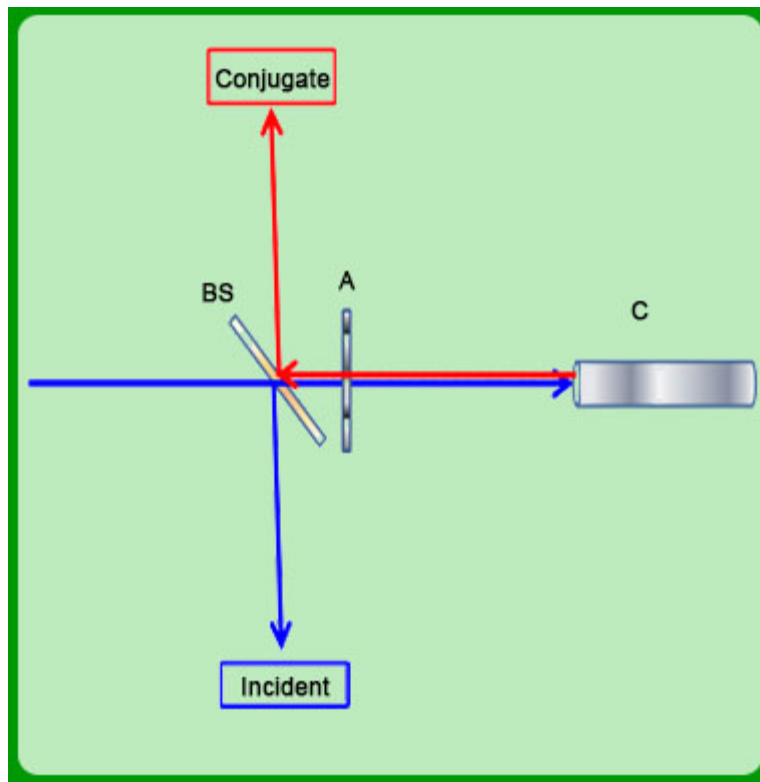


Figure 25.1

The beam splitter BS serves to record the input and output beam profiles separately. A is a ground glass plate used to distort the beams passing through it. The 1m long cell C contained methane gas at 125 atmospheric pressure. They observed that refracted SBS wave after passing through the distorted plate A had almost the same spatial quality as input laser beam before it was distorted by A. Thus the reflected SBS wave front was such that it produced an original wave front on passing through the aberrating glass A.

This would happen if the output SBS beam had a wavefront which was a replica of that of the pump beam but travelling in opposite direction. As we shall see this is equivalent to saying that the output wave amplitude is proportional to the complex conjugate of the input wave amplitude. The process of such oppositely travelling replica wave fronts is called optical phase conjugation.

This amazing discovery of the method of wavefront correction did not receive any great attention immediately although Nosach et al[2], from the same group, had demonstrated that the method can be

used to generate good quality amplified beams from relatively poor quality (inhomogeneous) laser amplifiers. SBS is an efficient process and has been studied a great deal as a method of optical phase conjugation. We note that the phase conjugate beam has a slightly shifted frequency compared to that of the incident one. References [3-13] provide reviews of this and other methods of nonlinear optical Phase conjugation (OPC).

A spurt in the study of optical conjugate came with the work of Hellwarth[14] who showed that the phase- conjugate beam can be obtained by degenerate four -wave -mixing (DFWM) also. Since then this process has been investigate vigorously as a means of optical phase conjugation as well as for many other applications. In the following we shall concentrate on DFWM as a method of obtaining phase-conjugation.

To understand the phenomenon of DFWM, consider two counter -propagating laser beams, called the forward (f) and backward (b) pump waves, respectively and a third wave called the probe wave (p), all having the same frequency ω intersecting in a nonlinear medium as shown in figure 25.2.

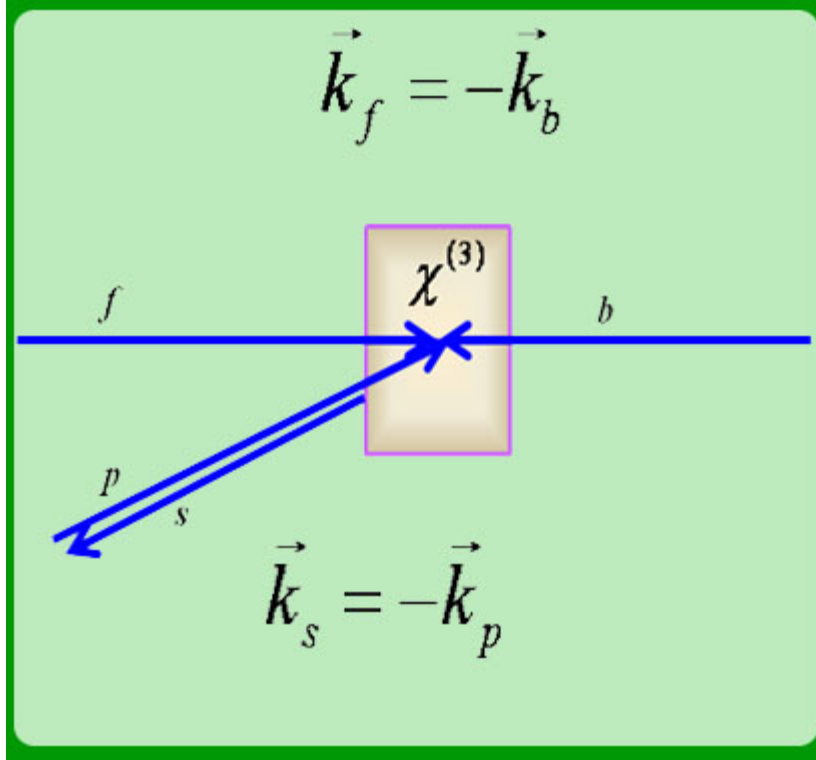


Figure 25.2

We look for a fourth wave also at ω generated as a result of this interaction. All the four waves -3 input and 1 output- have the same frequency (hence the nomenclature degenerate four -wave -mixing) and are distinguished from each other by their propagation vectors \vec{k}_f , \vec{k}_b and \vec{k}_p , respectively for the input waves and \vec{k}_s for the output or signal wave. As in the standard theory of nonlinear optical wave propagation, let the electric fields E_i in each input wave (i=f, b or p) be written as

$$E_i(\vec{r}, t) = A_i \exp \left\{ i \left(\vec{k}_i \cdot \vec{r} - \omega t \right) \right\} + \text{c.c.} \quad (25.1)$$

Each of the interacting fields satisfies

$$\nabla^2 E_i - \frac{\epsilon}{c^2} \frac{\partial^2 E_i}{\partial t^2} = \mu_0 \frac{\partial^2}{\partial t^2} P_i^{NLS}(\omega) \quad (25.2)$$

where ϵ is the appropriate dielectric function at frequency ω .

Interacting fields in the medium are coupled by the third order nonlinear susceptibility

$\chi^{(3)}(-\omega, \omega, -\omega, \omega) \equiv \chi^{(3)}$. These induce a nonlinear polarization which oscillates at their mixed frequency: $\omega_s = \omega - \omega + \omega \equiv \omega$. This nonlinear source polarization $P^{NLS}(\omega)$ is given by

(By geometry of interaction $\vec{k}_f = -\vec{k}_b$)

The spatial dependence of the nonlinear source polarization enables it to emit the phase matched signal field: $\vec{k}_s = -\vec{k}_p$. Thus the signal wave counter propagates the incident probe wave.

$$\begin{aligned} P^{NLS}(\omega) &\propto \epsilon_0 \chi^{(3)} E_f E_b E_p^* \\ &\propto \epsilon_0 \chi^{(3)} A_f(\omega) A_b(\omega) A_p^* \exp(i(\vec{k}_f + \vec{k}_b - \vec{k}_p) \cdot \vec{r}) \\ &\epsilon_0 \chi^{(3)} A_f(\omega) A_b(\omega) A_p^* \exp(-i(\vec{k}_p) \cdot \vec{r}) \end{aligned} \quad (25.3)$$

(Since by geometry of interaction $\vec{k}_f = -\vec{k}_b$)

The spatial dependence of the nonlinear source polarization enables it to emit the phase matched signal field: $\vec{k}_s = -\vec{k}_p$. Thus the signal wave counter propagates the incident probe wave.

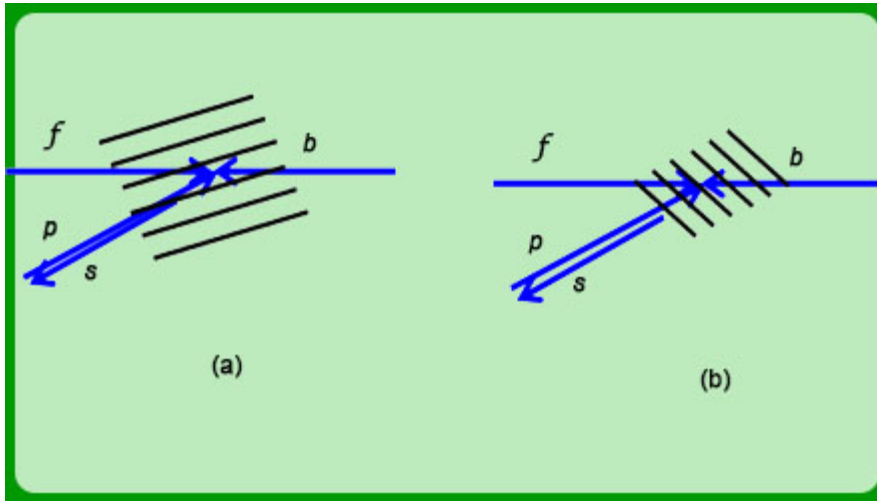


Figure 25.3

Alternatively, signal generation in DFWM can be understood in the following simple picture. Interference of the forward pump and probe beams produces a spatially periodic intensity modulation in the nonlinear medium (see figure 25.3(a)). Consequently, a replica of the intensity modulation is casted in optical properties e.g. refraction or absorption by nonlinear interaction. A spatially periodic pattern of optical properties behaves as a grating which diffracts the backward pump beam. The Bragg diffraction condition is satisfied such that the diffracted beam counter propagates the probe beam. Similarly, grating produced by the probe and backward pump beam diffracts the forward pump again to propagate opposite to the probe beam (see figure 25.3(b)). We will discuss the induced grating aspect of the process in more detail little later.

DFWM process can be formulated using the theory outlined in lecture#19. Total field in the medium can be written as

$$E_{total} = E_f + E_b + E_p + E_s \quad (25.4)$$

This field produces the nonlinear polarization for each of the interacting field. We assume that $\chi^{(3)}$ for the medium is real and the two pump beams have equal intensities which remain constant during the interaction. It can be shown that the product of the amplitudes of the forward and backward fields is spatially invariant under these conditions. We will, therefore, consider the spatial evolution of the probe and signal fields only. The source polarizations for the two fields for our intrinsically phase matched geometry can be written as

$$P_p^{NL} = 6\epsilon_0 \chi^{(3)} (|A_f|^2 A_p + |A_b|^2 A_p + A_f A_f A_p^*) \quad (25.5)$$

$$P_s^{NL} = 6\epsilon_0 \chi^{(3)} (|A_f|^2 A_s + |A_b|^2 A_s + A_f A_f A_s^*) \quad (25.6)$$

Following the general theory of four-wave-mixing, we can write the spatial evolution equations for the probe and signal field in SVEA as

$$\frac{dA_p}{dz} = i\kappa' A_p + i\kappa A_s^* \quad (25.7)$$

and

$$\frac{dA_s}{dz} = -i\kappa' A_s - i\kappa A_p^* \quad (25.8)$$

where

$$\kappa' = \frac{3\omega}{nc} \chi^{(3)} (|A_f|^2 + |A_b|^2) \quad (25.9)$$

$$\kappa = \frac{3\omega}{nc} \chi^{(3)} A_f A_b \quad (25.10)$$

Using the variable transformation

$$A_p = A_p' e^{i\kappa' z} \quad (25.11)$$

and

$$A_s = A_s' e^{-i\kappa' z} \quad (25.12)$$

We can write the equations (25.7) and (25.8) as

$$\frac{dA_p'}{dz} = i\kappa A_s' \quad (25.13)$$

and

$$\frac{dA_s'}{dz} = -i\kappa A_p' \quad (25.14)$$

One can eliminate the coupled nature of these equations by differentiating w.r.t. z

$$\frac{d^2 A_s'}{dz^2} + |\kappa|^2 A_s' = 0 \quad (25.15)$$

We can readily write its solution in the form

$$A_s'(z) = B \sin |\kappa| z + C \cos |\kappa| z \quad (25.16)$$

Constants B and C are to be determined from the boundary conditions. If we assume that there is signal field is injected into the medium at $z=L$ i.e. $A_s'(L) = 0$, then one can write the solutions for the probe and signal field.

$$A_p'(L) = \frac{A_p^{*}(0)}{\cos |\kappa| L} \quad (25.17)$$

and

$$A_s'(0) = \frac{i\kappa A_p^{*}(0)}{|\kappa|} \tan(|\kappa| L) \quad (25.18)$$

Note that

1. The transmitted probe wave is more intense than the incident one.
2. Also, the signal field can have intensity from zero to infinity and depends upon the value of $|k|L$

which involves $\chi^{(3)}$. Thus if the interacting field is scanned across an internal resonance of the medium, one will see very large signal enhancement at the resonance. Thus one can perform ultrahigh resolution spectroscopy as the resolution is only limited by the laser line width. This is also attractive as the signal is well directed which circumvents issue of signal collection efficiency. For ultrahigh resolution spectroscopy one actually keeps the pump beam frequencies fixed near the resonance and scans the probe across the resonance. A resonantly enhanced signal is generated only when the probe is tuned to the material resonance. Since all beams have very nearly same frequencies, this variant is called the nearly degenerate-four-wave-mixing(NDFWM). The signal generated is still well directed.

Wavefront recovery:

It is clear from equation 25.18 that in the $z = 0$ plane the signal wave amplitude is proportional to the complex conjugate of the probe amplitude.

From this it can be shown [4] in the scalar wave theory that the signal wave amplitude is the complex conjugate of the probe wave amplitude through the half- space $z \leq 0$. Thus for a probe wave described by

$$E_p(r, t) = E_p \exp\{-i\phi(x, y)\} \exp\{-i(\omega t - kz)\} + \text{c.c.} \quad (25.19)$$

The signal wave is given by

$$E_s(r, t) = E_s \exp\{-i\phi(x, y)\} \exp\{-i(\omega t + kz)\} + \text{c.c.} \quad (25.20)$$

This can also be written as

$$E_s(r, t) = E_s \exp\{i\phi(x, y)\} \exp\{i(\omega t + kz)\} + \text{c.c.} \quad (25.21)$$

Thus, apart from a multiplicative factor, the signal wave is a time reversed replica of the incident probe wave, i.e., at every given $z \leq 0$ the incident wave and the conjugate wave have the same wave front although the two directions of propagation are mutually opposite.

Even when this signal wave passes through a linear phase-distorting medium e.g. an aberrating glass plate, this property of time reversal is maintained due to the time reversal invariance of the wave equation so long as scattering is negligible. Hence the final output beam has the same wavefront as the incident one.

In terms of geometrical optics one can say that a phase conjugate mirror reflects each and every incident ray back onto itself. Thus converging rays diverge on reflection by a phase conjugate mirror and rays radiating from a point source would always focus back to the point no matter what linear and transparent elements are put between the phase conjugate mirror (PCM) and the source. That this is so is easily seen in the DFWM scheme where we note that because of the phase- matching condition the generated beam travels back along the probe irrespective of the angle of incident of the probe beam. Figure 25.4 illustrates how this ability to send every ray back on itself helps undo or heal the distortion caused by an inhomogeneous transparent medium. Parallel incident rays are refracted into different direction by an aberrator A—an irregular shape or inhomogeneous transparent object. On reflection by a normal mirror M the direction of the various reflected rays are determined by the corresponding angle of incidence at the mirror. Only some of these rays will pass through A on the return path and even these will not emerge parallel to each other. On the other hand a phase conjugate mirror (PCM) reflects each ray back on itself. The reflected rays thus retrace their paths through the aberrator and emerge parallel from the aberrator. We should note, however, that we assumed that the aberrator would preserve the polarization purity of the beam.

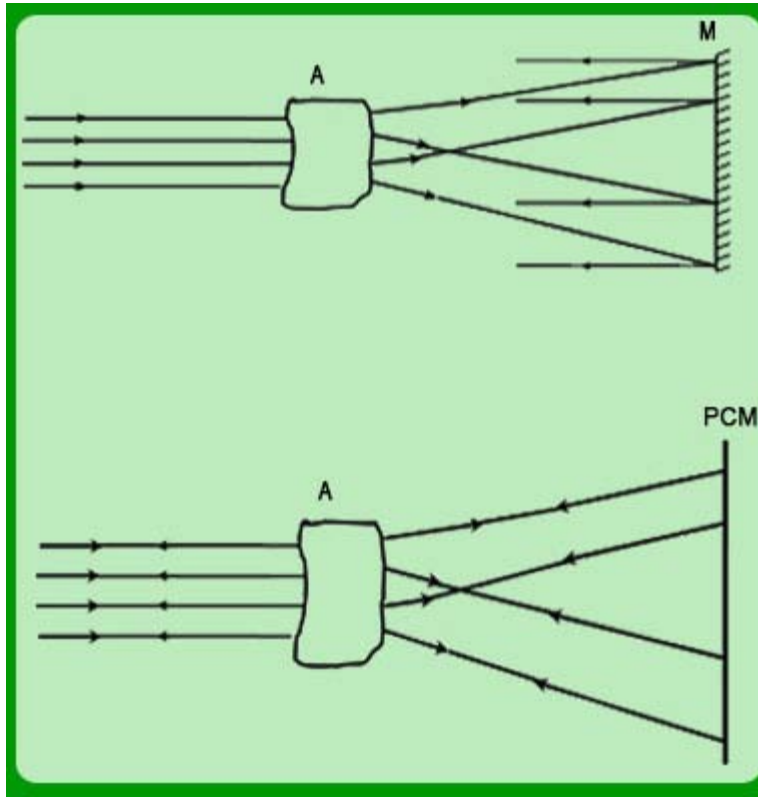


Figure 25.4

Since usually an inhomogeneous medium will cause polarization scrambling, considerable effort has been devoted[15] to evolve OPC scheme which reverse the wavefront for an arbitrarily polarized probe beam. Also, we have neglected back scattering by the aberrator[16].

This brief discussion already shows that a phase conjugate mirror is a fundamentally new optical element and opens up revolutionary possibilities. We discussed some of them in the following. Of course, some properties of a phase conjugate mirror such as reflectivity, polarization and spectral response depend on the nonlinear optical material used for the process.

DFWM as real time holography:

Degenerate four –wave- mixing is sometimes called real time holography. To see this connection let us note that for isotropic media the nonlinear sources polarization can be written as [8]

$$P^{NLS}(\omega) = A(\theta)(E_f \cdot E_p^*)E_b + A(\pi - \theta)(E_b \cdot E_p^*)E_f + (E_f \cdot E_p)E_p^* \quad (25.22)$$

Where θ is the angle between the f and p beam. The first two terms here have simple holographic analogues as can be seen from the following consideration. if we look at the intensity pattern created by the simultaneous presence of two beam f and p we will find a stationary interference pattern depicted by

$\text{Re}(E_f \cdot E_p^*)$ Superposed on a constant background proportional to $|E_f|^2 + |E_p|^2$ the intensity-dependence of the refractive index of the medium converts this intensity pattern into a refractive index pattern or a volume hologram.

If f and p both are plane waves, the interference pattern is a grating with period $2\pi/|\vec{k}_f - \vec{k}_p|$. The third beam b can now diffract from this volume phase grating and following the Bragg condition create a beam directed apposite to the probe beam. Similarly a p-b grating diffracts the forward pump beam. The connection between DFWM and holography is now quite obvious. The object wave in holography is replaced by probe beam, the reference wave by one of the two pump beam and the readout beam by the second pump beam. The difference is that recording is not permanent and reading may be done simultaneously. The second difference of course is that in DFWM one does not require to develop the hologram. Most of the tricks of image manipulation used in holography can be adopted and get information processed in real time. Clearly, for the volume hologram to exist any modification of the complex refractive index would do; we thus have a phase hologram when only the real part of the refractive index depends on the intensity and an absorption hologram when the absorption is intensity dependent. The third term in equation 26.11 is not interpretable in term of a temporally stationary

grating. Unlike the other two terms. It occurs even if the probe beam is polarized perpendicular to both the pump beam and is due to coherent interaction among the three beams. When the frequency as well ω as 2ω is away from all the resonances of the medium the three terms in equation 25.22 are expected to be of comparable magnitude. However, when, ω is closed to a resonance frequency of the medium, the grating contributions dominate. In the case of the resonant excitation real transfer of energy occurs from the laser to the medium. The intensity pattern created by the interference of the two “writing” beam is transferred into an “excitation” pattern. The excited medium has a different complex dielectric function compared to the unexcited state.

The change in the real part corresponds to the phase- modulating volume grating and that in the imaginary part to absorption modulation grating.

In this situation the time-dependence of the DFWM process is characterized by at least three important time constants. The response time or the rise time is the time constant characterized the establishment of the grating. The second time constant- the recovery time – characterizes the fall time of the nonlinear polarization in the medium. In a nonresonant situation the above two times are expected to be the same and determined by the frequency of the dominant virtual excitation. However when real excitation occurs in the medium, these two times can be very different. Moreover, a third time constant becomes important if the medium is homogeneous and the excitation can move through the sample. The ability of the excitation to move implies that the spatial variation of the excitation can vanish even before de-excitation of the medium. The corresponding characteristic time is called **the diffusion time or the grating washout time**.

For illustration let us consider the case of a semiconductor excited by a laser with photon energy $\hbar\omega \geq E_g$, the band gap. The excitation in this case leads to the generation of electron-hole pairs. An electron-hole pair may diffuse before it can recombine. The diffusion of the excited state implies a washing out of a sinusoidal grating in a time of the order of $\tau_D = \Lambda^2 / 4\pi^2 D_a$ where D_a is the ambipolar diffusion coefficient and Λ is the period of the grating[17]. It is important to note at this stage that the three terms in equation 25.22 can be separated by choosing appropriate polarization of the three input beams. If, for example, the forward pump and probe beams are polarized parallel to each other and the backward pump is polarized perpendicular to both, only the f-p grating is formed and the signal beam being due to the scattering of the backward pump beam from this grating is polarized perpendicular to the probe beam.

Using pulsed laser the two grating terms can also be distinguished from each other by delay one of two pump pulses with respect to the probe.

In a typical experimental set-up for efficient phase conjugation the probe beam makes a small angle with respect to one of the pump beams say the forward beam so that the interaction volume is reasonably large. Then the periodicities of the two grating (b-p and f-p) are rather different. For $\theta \ll 1$

a quick estimate gives the periods of the two gratings as $\lambda/2n$ and $\lambda/n\theta$ where λ is the wavelength of the laser radiation in the medium and θ is the angle between the probe and the forward pump beam also in the medium. In this type of situation, the f-p grating is also the slower to disappear if diffusion time τ_D is much smaller than the recombination time τ_R consequently the response of the f-p grating is much greater than of the b-p grating. It is pertinent to note that from this viewpoint of transient hologram DFWM has been studied in semiconductor [18] and saturable absorber [19] even before the work of Hellwarth [14]. In fact, Woerdman[18] had also noted that the disappearance of the grating is primarily due to ambipolar diffusion of electron-hole pairs. The method has often been used in investigation of detection-hole diffusion in semiconductors [19, 20]. A very interesting perspective on laser-induced grating in a variety of situation is given by Eichler [19].

Imaging application:

The fact that a PCM reflects every ray on to itself endows it with the capacity of image projection with large numerical aperture. Detailed studied [12,34] by Levenson and colleagues have demonstrated the feasibility of developing a high resolution, large-numerical-aperture optical lithography set-up based on PCM. In the early experiments on image projection a low power continuous wave Argon ion laser was used with photorefractive BaTiO₃ as the nonlinear medium. The exposure times required several hours. Later experiments were performed with the third harmonic of a Q-switched Nd:YAG laser and R6G dye solution was used as nonlinear medium [34]. The mechanism of nonlinearity in this case involves the temperature- dependence of the refractive index. Briefly, the pump-probe interference pattern is converted by absorption and subsequent thermalization into a temperature grating.

The temperature-dependence of the refractive index converts that into a phase-modulating grating. The

reflectivity of this kind of phase conjugate mirror has been demonstrated to be in excess of 300%. It should be noted that uv lasers are the most effective for photolithography application because of the rich variety of photochemical reaction in the uv region and because the shorter the wavelength, the higher the resolution achievable.

Another kind of imaging application of DFWM is by virtue of directional output that the process shares with the well-known coherent anti-Stokes Raman scattering (CARS) which has been used extensively for spatially resolved spectroscopic studies of highly luminescent objects such as flames and plasmas. For example, Ewart et-al [35] have used DFWM to image atomic concentration in plane cross-section of a flame.

The two dye laser pumps are focused cylindrically so that 90% of the pump energy is contained in a thin sheet (-25 mm x 0.5 mm). The probe beam illuminates a circular cross-section. The DFWM signal images the distribution of the sodium atoms in the flame with a resolution of up to 100 μm . This application of DFWM spectroscopy is also has significant potential [36]. The method is complementary to CARS, since in CARS vibrational excitation in molecules are more conveniently probed while in DFWM electronic excitation in atom or small molecules are more conveniently probed.

DFWM and optical computing:

There are several interesting applications of DFWM in optical computing. As already noted, DFWM can be viewed as real time holography. So it allows one to do holographic optical image processing in real time. This has been discussed in several reviews [4-7].

Second, DFWM can be exploited [37] in implementing the geometrical shadow casting methods [38, 39] for digital optical logic. In this method the logical input A and B are in the form of coded, geometrical patterns. For example, A is represented by horizontal binary pattern and B by a vertical binary pattern as shown in figure 25.5(a). The logic function to be implemented is represented by a 4-cell decoding patterns such as the one shown in figure 25.5(b) for XOR function. Decoding patterns for other logic function are given by Yatagai [39].

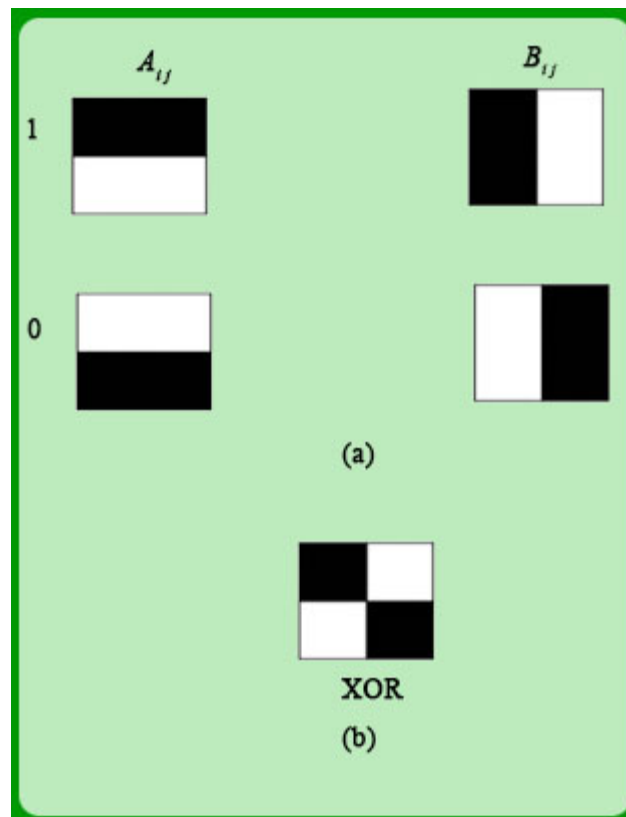


Figure 25.5

In the output, logical 1 is the situation when light passes through any of the four cell of the decoding mask superposed with the input masks A and B. A completely dark output field or zero transmission is taken as the logical zero. If this logic is implemented by mechanically superposing the three transparencies- the two input pattern and one decoding pattern- the advantage of parallelism of optical computing is retained but the processing speed i.e. the number of operation performed per second, is obviously very slow. This can be improved greatly if the transparencies are replaced by electrically addressed or optically addressed spatial light modulators [40]. The same process can be performed

through DFWM also since the DFWM output is proportional to three inputs which can now be coded into the two pump beams and the probe beam as shown in figure 25.6.

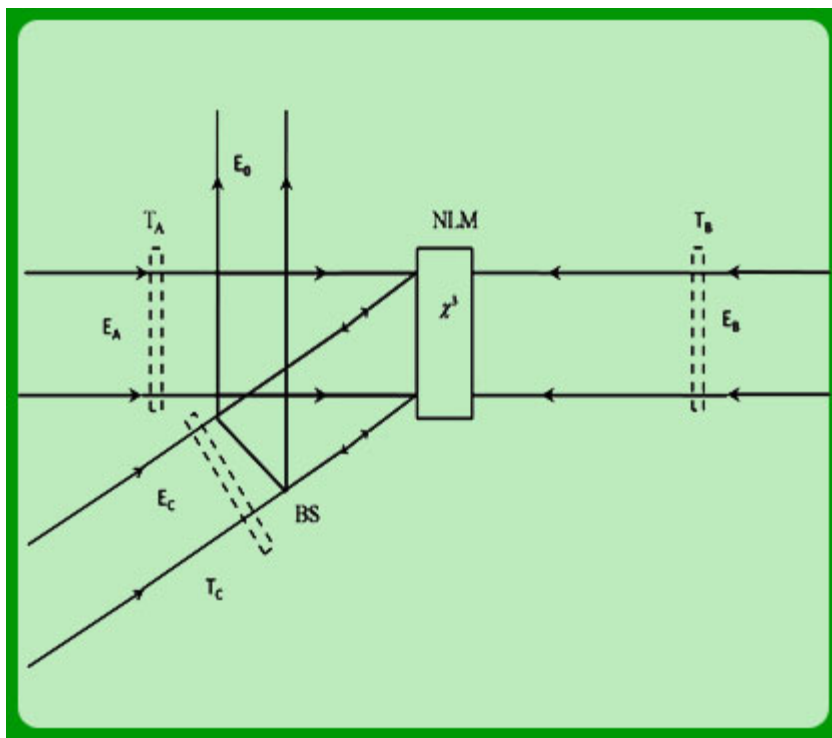


Figure 25.6

The signal beam carries the logical output information.

The main advantage of using DFWM is the several logic functions can be implemented simultaneously by using different probe beam as illustrated in figure 25.7 for two functions. Further, since DFWM is a coherent process, the method can be combined with the usual coherent analogue techniques [41].

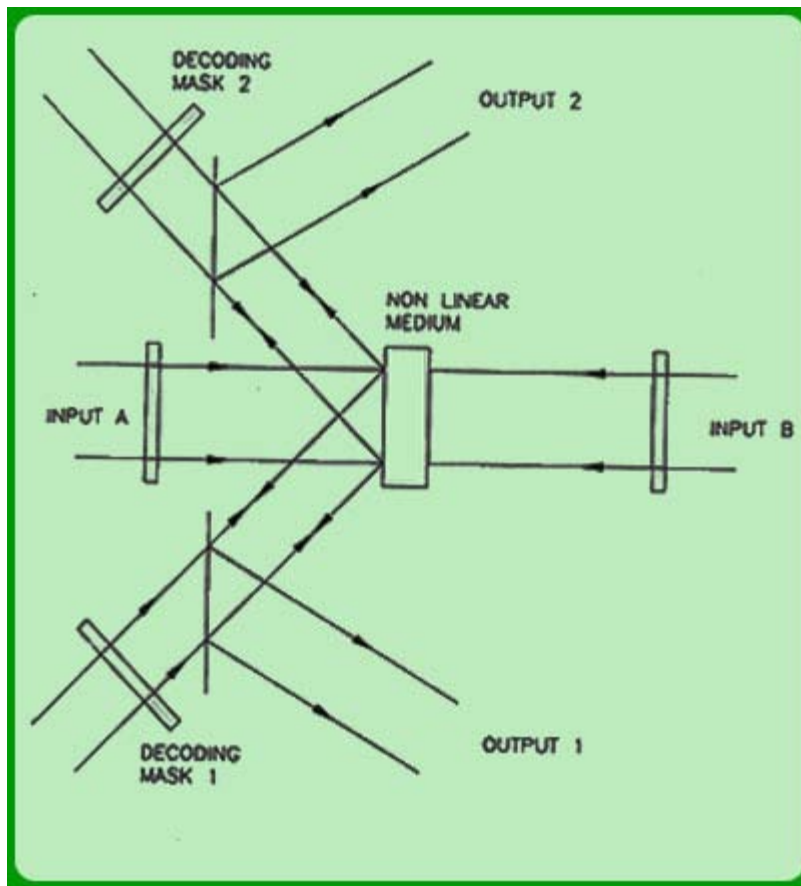


Figure 25.7

Yet another promising optical computing application of OPC is in the realization of optical associative memories [42]. In present day digital electronic computers the information is stored in random access memories (RAM) in which each memory registered is identified by a number. To read or call the information stored in any given register is simple in such an arrangement. However, if we want to find full information about an item about which we possess part information we have to scan each memory register one by one to check by comparing if the item stored there matches the part information. In contrast, the human brain can recall or recognize people by their faces or their dress or the way they walk, each piece of information is obviously a small part of the information we have about that person. An information storage system which permits direct recall or reconstruction of the complete information about an item from the knowledge of one part of that information is called an associative or content addressable memory. In one particular realization of such a memory one can recall the complete image of an object when any part image is used as input [42]. Volume holograms of several images are first recorded sequentially in the same hologram using a distinct reference beam for each object wave front.

The reference beams are all plane waves of the same frequency but with different directions of incidence. Let $\{a_i\}$ be the object waves recorded with b_i denoting the plane wavefront used to record the i th hologram. Suppose now we want to reconstruct the wavefront a_1 from a wavefront \hat{a}_1 which carries part of image depicted by a_1 . The arrangement proposed by Soffer *et al* [43] to do this is shown schematically in figure 25.8.

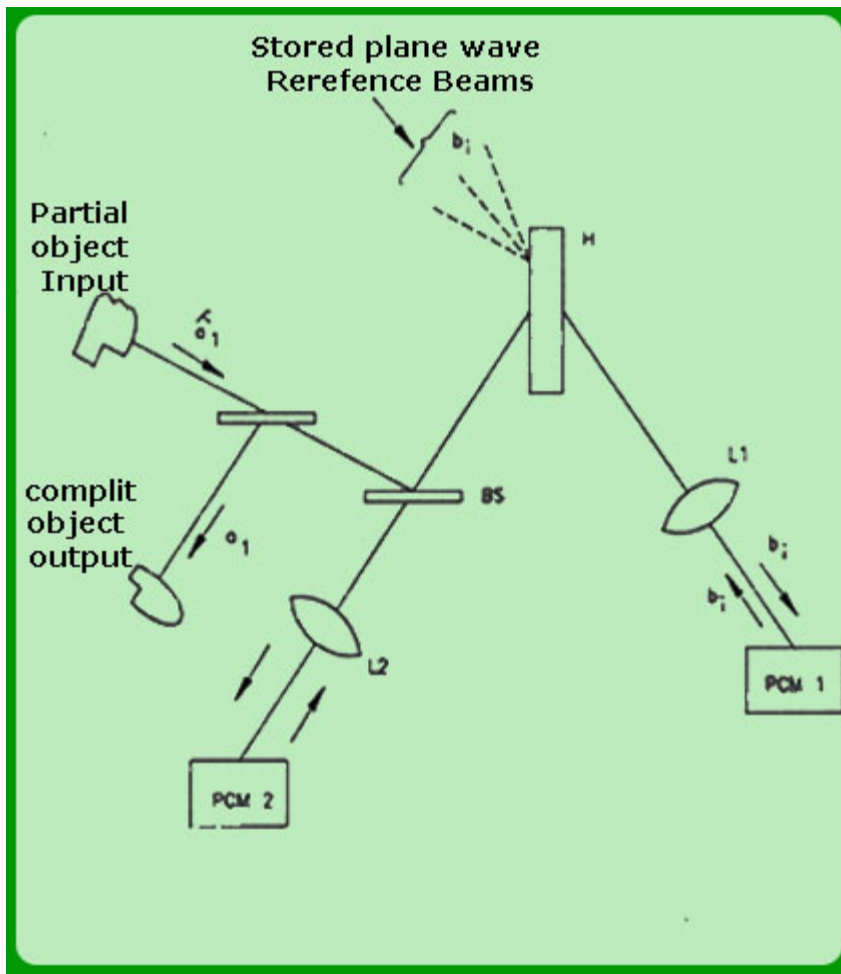


Figure 25.8

H is the composite hologram mentioned above which contain the interference pattern of wavefront $\{a_i\}$ with corresponding plane wave $\{b_i\}$, PCM_1 is a thresholding phase conjugate mirror whose reflectivity deepedns on the input intensity. This could be a PCM based on self-pumped DFWM [44] or on SBS.

The second conjugate PCM_2 is an externally pumped phase conjugate mirror whose reflectivity is dependent on the input intensity. The partial wavefront a_1 on diffraction by hologram H produces the corresponding plane wave b_1 with some distortion. On being focused (Fourier transformed) by lens (L_1) the plane wave component of the diffracted beam produces a higher intensity image.

Because of the intensity- dependent reflectivity of PCM_1 , this component is preferentially generated in the phase- conjugate reflected beam.

On being Fourier transformed by the lens L_1 this produces a relatively less distorted plane wave b_1 . This when diffracted by H produces a better approximation to the complete wavefront a_1 which on being reflected by PCM_2 will start the next cycle. Obviously there will be diffraction losses in each cycle as well as some other losses. Thus the complete object wavefront will be generated without distortion after many cycles provided the gain in PCM_1 and PCM_2 is sufficient to overcome these losses. We should also note that, by the very nature of associative memories, if the partial information has substantial overlap with several images, the arrangement could produce any one of them or may not converge to anyone. There are many other applications of DFWM in optical computing. Of these schemes those in which the polarization of the beam is used to carry the information are of particular interest because of their easy cascability. For more detail we refer the reader to some representative papers [45,46].

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Recap

In this lecture you have learnt the following

- An outline of the physical phenomena involved
- Theory of degenerate four-wave-mixing (DFWM) and Optical phase conjugation
- Equivalence of DFWM to real time holography.
- Applications in
 - Wavefront recovery,
 - Imaging and
 - Optical computing