Interferometry with a novel composite material

R. Krishna Mohan, S. Balan, P. S. Narayanan and
C. K. Subramanian

Department of Physics, Indian Institute of Science, Bangalore 560 012, India.

An interesting application of optical phase conjugation is phase conjugate interferometry. We report here a new approach to real-time interferometry which combines the high phase conjugate efficiency of photorefractive crystals and the fast response times offered by dye-doped polymer films by using a composite structure. The ability of this material to generate two independent but overlapping phase conjugate waves, which can interfere to reveal the phase changes in a test object, is discussed and demonstrated with a specific example.

PHASE conjugate interferometry (PCI) is an important application of optical phase conjugation (OPC). It has been demonstrated in a variety of ways that any non-uniform phase change caused by atmospheric turbulence or the distortions due to optical elements can be corrected for due to the 'healing' property of a phase conjugate mirror (PCM)[1]. The 'auto-tracking' property of the phase conjugate wave and a possible amplification with high gain phase conjugators is also widely known. Hence an interferometer in which the conventional mirror is replaced by a PCM carries several advantages over the conventional interferometric configurations[2].

Laser-induced gratings have been observed in several nonlinear media. Ferroelectric photorefractive crystals have been shown to be capable of generating a refractive index modulation, through the creation of a space charge field and a subsequent Pockel's effect, when a modulated intensity pattern is formed in them. The modulated refractive index pattern acts as a grating for creating new optical fields through diffraction. The ability of these crystals to retain the recorded hologram for a finite time has been exploited for interferometry. Photorefractive crystals like $BaTiO_3$, BSO have shown high recording-erasure sensitivity and have been used to realize real-time, time average and double exposure interferometry[3,4].

Dye-doped films on the other hand have been shown to be suitable for PCI even at low irradiance levels[5]. The dye-doped polymer films like erythrosin–PVA and methylred–PMMA can generate phase conjugate (PC) waves simultaneously by degenerate four wave mixing (DFWM) with a fast response time and a holographic process with a slower response. The fast DFWM signal is due to the saturation of absorption of the dye medium. The time scales of the recording and erasure of grating in these media due to this mechanism is governed by the

singlet–singlet transition probability and the triplet relaxation processes. The holographic grating formation is due to irreversible photoisomerization of the dye. The grating formation and decay times are much slower than the saturation of absorption since rotation about C–C bonds and the photoreduction of dye molecules are involved. This difference in the response times between the two mechanisms was exploited and by choosing a proper FWM geometry both real-time and double exposure PCI were realized[6,7].

A comparison of the values of PC reflectivities in various media reveals that photorefractive crystals are about two orders of magnitude more efficient than dye-doped thin films. But the response to optical fields of the photorefractive effect is generally two orders of magnitude slower than the saturable absorption contribution in dye films. Here we report a new approach which combines the high diffraction efficiency of a photorefractive crystal and the fast response times offered by dye-doped polymer films.

The novel approach is to prepare a composite material containing both a photorefractive crystal and a dye-doped film. We have prepared a new composite material by depositing a thin layer of an absorbing dye-doped polymer on a photorefractive substrate. The substrate chosen by us is a thin ferroelectric $LiNbO_3$ crystal on which an Aniline Yellow-PMMA film is deposited.

This composite material can now generate two kinds of PC waves in a DFWM geometry. The crystal will generate a PC signal due to the creation of a space charge field which modulates the refractive index of the crystal which then acts as a grating. The diffraction efficiency of this grating quickly reaches a steady state value and PC intensity saturates. The dye film can, in the same geometry, also generate a PC wave due to saturation of absorption of the dye. The growth of the PC signal due to the dye film is almost instantaneous in comparison. This PC signal decays within a few milliseconds if any one of the three beams is turned off. No permanent photochemical change is observed. The difference in the time scales of the two PC waves is made use of in PCI with this composite material.

The experimental setup used for performing PCI is shown in Figure 1. The 488 nm radiation from a CW Argon ion laser is split into three beams which are made to overlap in the composite material in a typical FWM geometry. The test object of interest, TO, is placed in the path of probe wave I_{PR}. A microscope objective MO is used to expand the probe beam which then passes through the test object. A collecting lens L ensures that maximum phase information of the test object is retained This probe beam which contains the phase information of the object is focused on the composite where two counterpropagating beams, forward propagating pump I_P and backward propagating read I_R beams overlap. A part of the PC signal generated by the composite material is reflected by the beam splitter BS_3
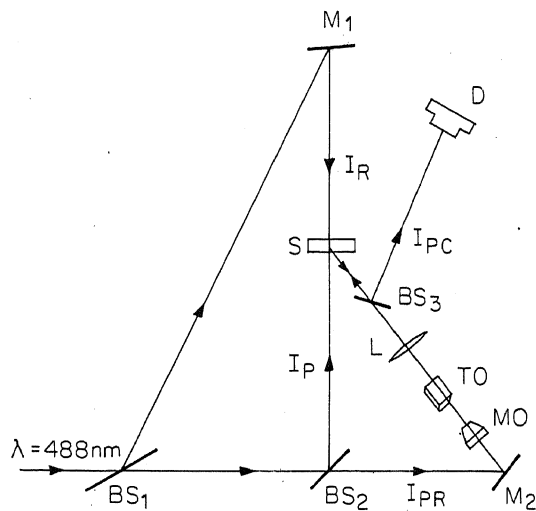
Figure 1. Experimental layout for phase conjugate interferometry. BS, Beam splitter; M, Mirror; D, Photographic plate.

Figure 2. Interferogram of a photographic transparency after a positional disturbance.

Figure 3. Interferogram showing thermal currents around a heater coil in a liquid.

onto a photographic plate for recording the interferograms.

The principle involved in the realization of PCI using this composite material is illustrated by taking some specific cases of interest. In the first case the test object chosen is a photographic transparency. The slide is kept in the path of the probe beam suitably expanded. Initially only pump and the probe beams are switched on. The hologram of the slide is recorded in the crystal in this two-beam coupling geometry. The holographic reconstruction can be done at any time later by turning the read beam on. No permanent holographic storage is observed in the dye film part of the composite structure. In the next step all the three beams are switched on simultaneously. A phase change in the object wave from the transparency is introduced either by way of a slight positional disturbance or an angular tilt in the optical elements. The dye film generates a new PC signal in this FWM geometry. Now two distinct images are formed by the read beam. One is due to the diffraction from the hologram written in the first step and the other is a FWM signal with the new object wave. These two PC waves carrying the two distinct images are travelling in the same direction and overlap in space. They are slightly different in phase and this shows up in the form of interference fringes as can be seen in Figure 2. The fine fringe pattern in the interferogram is distinctly visible and is due to a small positional disturbance of the test object. The fringe spacing is a measure of the phase changes. This is a double exposure type of PCI.

Real-time PCI has also been realized using the composite material. The refractive index changes due to thermal currents in a liquid is an example chosen for this purpose. A heater coil immersed in a bath of glycerine has been chosen as the test object and is placed in the path of the probe beam. In the first step the hologram of
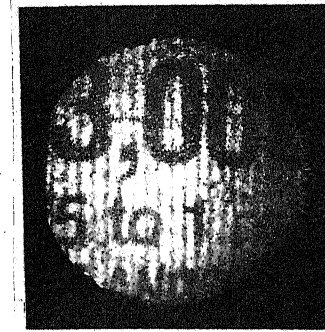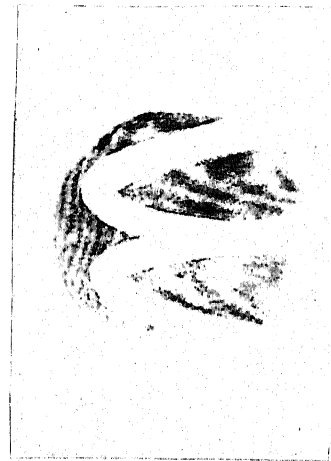
the coil immersed in glycerine is recorded in the photorefractive substrate of the composite through two-wave mixing. As soon as the heater is switched on, the refractive index of the liquid around the coil changes due to the heating. This introduces a phase change in the object wave. In the second step, in a FWM configuration, the dye film generates a new PC signal which contains the new phase information of the object. Since the response of the dye film is of the order of milliseconds the phase changes in the object wave on these time scales can be continuously monitored in the form of interference fringe pattern. Figure 3 shows a typical interferogram of the thermal currents around the heater coil after it is switched on. Since the phase changes are due to the gradients set up by the thermal currents, a real-time study of the interference fringes reveals the heat transport properties of the medium.

The practical utility of interferometry in the field of non-destructive testing is widely known. Both real-time

and double exposure holographic interferometry have been extensively used in the visualization of structural deformations and vibrations. The conventional techniques using a two-beam geometry involves the separation of the transmitted object beam and the image-bearing diffracted beam. So the quality of the interference pattern depends heavily on the transmission of the hologram and the suppression of the background intensity. The four-wave mixing (FWM) geometry employed by us helps in obtaining a PC image of the object which is free from distortions because of its aberration compensation property. The importance of the present work lies in the fact that by preparing a nonlinear medium, in the way suggested by us, one can attain a contrast of unity.

A new approach to combine the efficiencies of a photorefractive crystal and the fast response times offered by dye thin film, is discussed. This novel composite material gives two phase conjugate signals differing in their growth and decay times. It has been exploited in a simple four-wave mixing geometry to perform phase conjugate interferometry. The technique is free from the phase change problems associated with dye thin films alone and a very high contrast interferogram can be obtained by properly choosing suitable material and geometric parameters.

1. Pepper, D. M., *Sci. Am.*, 1989, **254**, 56–65.
2. Hopf, F. A., *J. Opt. Soc. Am.*, 1980, **70**, 1320–1323.
3. Feinberg, J., *Opt. Lett.*, 1983, **8**, 569–571.
4. Huignard, J. P.and Marrakchi, A., *Opt. Lett.*, 1981, **6**, 622–624.
5. Bar-Joseph, I. *et al*, *Opt. Lett.*, 1981, **6**, 414–416.
6. Nakagawa, K. and Fujiwara, H., *Opt. Commun.*, 1989, **70**, 73–76.
7. Nakagawa, K. and Fujiwara, H., *SPIE*, 1989, **954**, 11–15.

Investigations of the fullerene hydride $C_{60}H_{36}$

A. Govindaraj

CSIR Center of Excellence in Chemistry, Indian Institute of Science, Bangalore 560 012, India

Under Birch reduction conditions, C_{60} predominantly forms $C_{60}H_{36}$. Spectroscopic results are consistent with the structure predicted by calculations, involving four benzene rings distributed tetrahedrally in a spheroidal framework.

SINCE the discovery of the fullerene, there has been considerable effort to study the chemistry of these fascinating molecules[1,2]. A variety of reactions have been carried out on C_{60} generally giving rise to a mixture of products. Birch reduction of C_{60} has been reported[3] to yield a mixture of $C_{60}H_{18}$ and $C_{60}H_{36}$, while controlled hydroboration[4] followed by protonolysis gives $C_{60}H_2$. Calculations have been carried out on $C_{60}H_{60}$, with isolated double bonds[5–8]. Considering that several of the derivatives studied so far conform to the general formula $C_{60}X_{60-6n}$ (X = H, Br, etc), it is tempting to speculate whether some of these derivatives consists of n distinct benzene rings distributed in the fullerene cage[9]. $C_{60}H_{36}$ seems to be the most promising candidate to adopt such a structure with four isolated benzenoid rings.

The preparation of fullerene was carried out by contact arc vapourization of graphite[10,11]. Separation and purification of C_{60} were accomplished by a simple filtration technique using a charcoal – silica-gel mixture[12]. $C_{60}H_{36}$ was obtained by Birch reduction of c_{60} as follows. To excess Li/liquid NH_3, a solution containing 30 mg of C_{60} and 5 ml of *tert*-butanol in 200 ml methyl cyclohexane was added and stirred vigorously. The temperature of the reaction mixture was maintained at minus –33°C for 6–7 h by using a liquid ammonia refluxing condensor. The product was quenched with excess of ammonium chloride, washed with water, dried over anhydrous $MgSO_4$ and passed through a silica column to remove possible amine addition products. The new product in TLC showed a single spot (iodine-active) with an R_f value of around 0.37 in 30% CH_2Cl_2/hexane. After removing the solvent, a white solid product was obtained. This product was characterized by FAB and EI mass spectrometry, FTIR, NMR and UV spectroscopy.

Although the FAB and EI mass spectra of the purified product of Birch reduction show a distribution of mass peaks, the main line is at $m/z = 756$ (Figure 1) corresponding to a hydride $C_{60}H_{36}$. There was no feature to indicate the presence of a NH_2 group in the FTIR spectrum (KBr pellet). The FTIR spectrum showed characteristic frequencies associated with aliphatic C–H
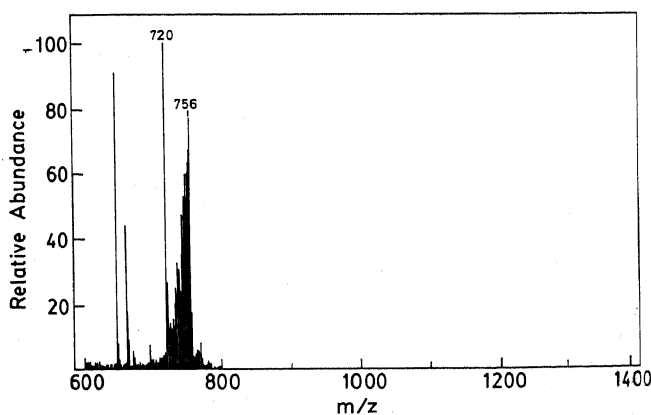

Figure 1. Electron impact mass spectra of $C_{60}H_{36}$.