Holographic interferometry of ultrafast transients by photochemical hole burning

A. Rebane

Physikalisches Institut der Universität Bayreuth, D-8580 Bayreuth, Federal Republic of Germany

J. Aaviksoo

Max-Planck-Institut für Festkörperforschung, D-7000 Stuttgart, Federal Republic of Germany

A novel approach for interferometric analysis of ultrafast transient processes as short as 10^{-13}–10^{-14} sec is suggested. The method of holographic storage of ultrafast time–space images in spectrally selective hole-burning materials is used. It is shown that differential interferometric comparison of time–space images, which are permanently stored, provides an effective means to resolve ultrafast transient signals in space and in the spectral domain.

Holographic interferometry has proven to be an effective method for the investigation of a variety of fast transient phenomena such as laser-induced plasmas and sparks and fast target displacements.[1] Momentary holographic images of the transient scenes, once stored in holograms, are further used to form interference patterns with reference images revealing minute changes in their optical and geometrical properties.

To carry out interferometric measurements of fast processes, laser pulses as short as 10^{-13}–10^{-14} sec are currently available.[2] Unfortunately conventional holographic image-storage techniques fail to accommodate such short recording times owing to the large spectral width of 10^2–10^4 cm^{-1} and the short coherence lengths of 10^{-2}–10^{-4} cm of ultrashort laser pulses.

Recently a novel holographic image-storage technique, time–space holography, was introduced.[3–7] For the recording of the holograms this method uses special materials doped with photochemically reactive dye molecules cooled to cryogenic temperatures. At these low temperatures the homogeneous absorption lines of individual molecular absorbers are as narrow as $v_H = 10^{-3}$–10^{-4} cm^{-1} [the so-called homogeneous zero-phonon lines (ZPL's)],[8] while in the sum ZPL's of all molecules still make up wide absorption bands with characteristic widths of 10^2–10^4 cm^{-1} (inhomogeneously broadened absorption bands). Irradiation of these materials with monochromatic light results in permanent bleaching of the absorption bands, particularly at the applied wavelength. It has been well established[9–12] that this effect, called photochemical hole burning (PHB), is due to the strong resonant absorption caused by impurity ZPL's (Q factor of the zero-phonon transition is ~10^7). The PHB effect occurs when the act of absorption triggers in the impurity center some kind of chemical reaction or molecular displacement that alters the ZPL of the dye molecule. Materials with very slow (years) degradation of PHB bleaching have been reported.[13]

In the method of time–space holography all spectral components of the light signal are recorded simultaneously as long as the spectral extent of the signal does not exceed the width of the inhomogeneous absorption band.[3,4] For the creation of reference signals, ultrashort laser pulses with a broad spectrum are applied. It has been demonstrated experimentally[6] that PHB holograms can record not only the spatial image but also the full temporal structure of the scene if the duration of the scene does not surpass the inverse value of the homogeneous ZPL width. It is important to note that the time-domain storage of ultrashort signals takes place because of the property of the PHB holograms to memorize phase and amplitude relations between the spectral components of the light signal.[3–7]

Here we are going to demonstrate that by using time–space holograms in spectrally selective PHB storage media, holographic interferometry is feasible even if sampling laser pulses as short as 10^{-13}–10^{-14} sec are used. The principal advantage of the present method arises from the ability of time–space holograms to record full-scale scenes rather than frames of spatial images. As a result the analysis of the interferograms reveals not only the spatial changes but also spectral and temporal changes of the scene that occurred on the ultrafast time scale.

Two schemes of interferometric measurement are analyzed. In the first scheme the readout of the interferogram is performed with the help of a tunable narrow-band cw laser. The information about the temporal changes is encoded in the spectral interference fringe pattern, which is obtained through spectral scanning of the cw laser. In the second scheme two time–space images are compared directly, and the difference between them is obtained in a time-domain measurement.

Consider a thin PHB hologram plate positioned in the (x, y) plane being illuminated at a time $t = 0$ by a plane wave-front reference pulse with a deltalike temporal profile. Let this pulse be applied to the recording hologram plate at a small incidence angle θ (Fig. 1).
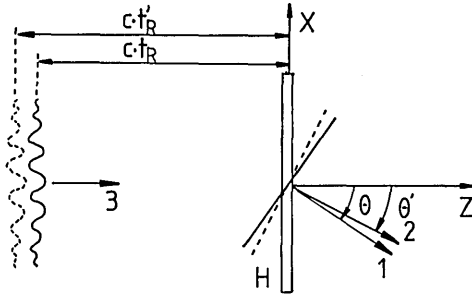
Fig. 1. Scheme for recording double-exposure time–space holographic interferograms. H, recording hologram plate; solid lines, wave fronts of the first-exposure pulses; dashed lines, wave fronts of the second-exposure pulses. Arrows 1 and 2 indicate the directions of the reference pulses in the first and second exposures; arrow 3 indicates the direction of both signal pulses.

The amplitude of the reference pulse in the plane of the hologram is given by

$$R(\mathbf{r}, t) = R_0 \delta(t + x\theta/c) \exp[i\omega_0(t + x\theta/c)], \quad (1)$$

where ω_0 is the optical carrier frequency.

Let the reference pulse be followed by a signal pulse with a short time delay t_R of duration t_s and with amplitude $S(\mathbf{r}, t)$ carrying the image of the scene under investigation:

$$S(\mathbf{r}, t) = s(x, y, t - t_R) \exp[i\omega_0(t - t_R)]. \quad (2)$$

In Refs. 5 and 6 it was shown that in order to record a time–space holographic image, the width of the inhomogeneous absorption band of the PHB medium has to be large enough to comprise all optical frequencies involved in the signal light pulse. Inhomogeneous widths of $\Delta\omega_G \simeq 10^{-2}$–$10^{-4}$ cm^{-1} are typical in PHB systems composed of organic dye molecules in polymeric host matrices. Following Refs. 5 and 6, let us here also suppose that the spectrum of the signal pulse does not extend beyond the limits of the inhomogeneous absorption band centered at ω_0. The minimum duration of the signal pulse can be estimated as $(c\Delta\omega_G)^{-1} \simeq 10^{-13}$–$10^{-14}$ sec.[14]

Let the delay and the duration of the signal pulse be small compared with the inverse value of the homogeneous ZPL linewidth of PHB molecules,

$$t_s < t_R \ll \nu_H^{-1}. \quad (3)$$

The combined action of the reference and signal pulses introduces permanent PHB bleaching of the hologram material. In a simple linear approximation[5,6] the PHB-induced changes of the amplitude transmission of the hologram can be expressed as linearly proportional to the light intensity applied,[15]

$$K^{(1)}(x, y, \omega) \sim (1 + i\hat{H})[I(x, y, \omega)]$$

$$= (1 + i\hat{H})[R_0^2 + |\bar{s}(x, y, \omega - \omega_0)|^2]$$

$$+ 2R_0\bar{s}(x, y, \omega - \omega_0)\exp[-i\omega(x\theta/c + t_R)], \quad (4)$$

where s stands for the Fourier image, and the Hilbert transformation

$$\hat{H}[f(\omega)] \equiv \frac{1}{\pi} \int f(\omega')d\omega'/(\omega' - \omega) \quad (5)$$

keeps the complex function $K^{(1)}(x, y, \omega)$ from violating the causality principle. Note that the second term in the hologram transmission function [relation (4)] contains full information about the image of the transient scene under investigation. The first term contains no holographic data and is omitted in what follows.

Now let the same hologram plate be used for a second exposure. In an analogy to conventional double-exposure interferometry techniques,[1] let the second signal pulse $S'(\mathbf{r}, t)$ carry the image of the same scene but under some different probing conditions. The amplitude of the second signal pulse is given by

$$\bar{s}'(x, y, \omega - \omega_0) = \bar{s}(x, y, \omega - \omega_0)\exp[i\varphi(x, y, \omega)], \quad (6)$$

where $\varphi(x, y, \omega)$ is the difference in optical path lengths between the signals S and S' at frequency ω. The reference pulse used for the second exposure is assumed to be identical to that in the first exposure with the exception of a slightly altered incidence angle $\theta' = \theta + \Delta\theta$. For the second exposure let us also introduce an additional delay $\tau \ll \nu_H^{-1}$ so that the time interval between the reference and the signal pulses becomes $t_R' = t_R + \tau$.

The amplitude transmission of the resulting double-exposure spectrally selective PHB hologram is given by

$$[K^{(1)}(x, y, \omega) + K^{(2)}(x, y, \omega)] \sim R_0\bar{s}'(x, y, \omega - \omega_0)$$

$$\times \exp[-i\omega(x\theta/c + t_R)] + R_0\bar{s}',(x, y, \omega - \omega_0)$$

$$\times \exp[-i\omega(x\theta'/c + t_R')]. \quad (7)$$

For the analysis of this holographic interferogram let us illuminate it with a plane wave-front narrowband cw laser beam of frequency Ω at an incidence angle of $\theta'' \approx \theta', \theta$. At the output of the hologram two diffracted mutually coherent monochromatic light signals will appear propagating at close angles. The intensity of this diffracted signal is given by

$$I(x, y, \Omega) \sim I_0(\Omega)R_0^2 |\bar{s}(x, y, \Omega - \omega_0)|^2$$

$$\times 2\{1 + \cos[\varphi(x, y, \Omega) - \alpha(x, y, \Omega)]\}, \quad (8)$$

where $I_0(\Omega)$ is the intensity of the readout beam and

$$\alpha(x, y, \Omega) \equiv \Omega x \Delta\theta/c + \Omega\tau. \quad (9)$$

The observed spatial distribution of the intensity [relation (8)] displays a set of interference fringes. The mean spatial period of the fringe pattern is determined by the small difference between the angles θ and θ'. The spatial disturbances of this reference pattern correspond, as in the case of ordinary holographic interferometry, to the alteration of the optical path length experienced by the interrogating laser pulses at the two different exposures.

By scanning of the laser frequency Ω, the wavelength dependence of the optical path length differences is obtained. It also reveals a second set of reference interference fringes coded for this time in the spectral domain. The mean spectral period of these

fringes is determined by the difference between the delay times t_R and t_R'. This provides a convenient reference in determining small optical path length variations within the spectral band of the interrogating pulses. Note that deviations from exact periodicity of the fringes in the spectral domain will indicate nonlinear phase modulation of the interrogating pulses by the transient process under investigation.

Furthermore, from the visibility of the spatial and spectral domain fringes the data on the amplitude changes of the signals $S(x, y, \omega)$ and $S'(x, y, \omega)$ can be extracted.

As a result exhaustive information is obtained about the difference between the two ultrafast transient scenes. Note that as a contrast to ordinary holographic interferometry, by processing the spectral data continuous temporal changes within the ultrafast scenes can be traced.

Now let the interferometric scheme considered above be modified so that the temporal order of the reference and signal pulses in the first exposure is reversed. In this case the linear transmittance function of the hologram after the first exposure is given by

$$K'(x, y, \omega) \sim R_0 \bar{s}^*(x, y, \omega - \omega_0)$$
$$\times \exp[-i\omega(-x\theta/c - |t_R|)]. \quad (10)$$

Let us perform the readout of this one-exposure hologram with the second signal pulse S' (or, which is equivalent, with its holographically stored replica). At the output of the hologram a signal diffracted at an angle $-\theta$ will appear. The time-domain amplitude of this signal is given by

$$A(x, y, \omega) \sim R_0[s'(x, y, t) * s(x, y, t)]$$
$$\times \exp[i\omega_0(t + x\theta/c - t_R)], \quad (11)$$

where $*$ denotes the time-domain correlation. Under certain conditions, discussed in detail in Refs. 16 and 17, the autocorrelation of the signal S displays a sharp correlation peak and can be substituted for in relation (11) by a delta function. In this case the time-dependent intensity observed at the output of the hologram is

$$|A(x, y, t)|^2 \sim |F^{-1}\{\exp[i\varphi(x, y, \omega)]\}|^2, \quad (12)$$

where F^{-1} stands for the inverse Fourier image.

By measuring the temporal and spatial structure of the intensity [relation (12)] directly in the time domain, the difference between the two transient optical signals $S(x, y, t)$ and $S'(x, y, t)$ is obtained.

In conclusion, we have demonstrated that holographic interferometric measurement techniques, when combined with permanent holographic storage in spectrally highly selective hole-burning media, provide detailed information about fast altering processes even if laser pulses as short as tens of femtoseconds are applied.

A. Rebane is also with the Bayreuther Institut für Makromolekülforschung. The permanent address of the authors is Institute of Physics, Estonian Academy of Science, 142 Riia Street, Tartu, USSR.

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