## Spectrally controlled interference of picosecond time-and-space-domain holograms

## Daniel Erni, Alexander Rebane, and Urs P. Wild

Physical Chemistry Laboratory, Swiss Federal institute of Technology, ETH-Zentrum, CH-8092 Zürich, Switzerland

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We show that two superimposed time-and-space-domain holograms in a persistent spectral hole-burning material can produce destructive interference, even if the path-length difference of the writing beams is constant. The cancellation of the amplitudes in the -1 diffraction order occurs when the two holograms have specially chosen spectral envelopes that represent a pair of complementary functions. By controlling the spectrum of the holograms we are able to perform coherent subtraction and addition of images.

Time-and-space-domain holography makes use of the spectral selectivity of materials with inhomogeneously broadened absorption bands to record (in addition to the spatial image) the spectral interference pattern of temporally nonoverlapping object and reference amplitudes.<sup>1–3</sup> In materials with persistent spectral hole burning the storage is established by a photochemical process that reduces the absorption of the media in proportion to the intensity distribution in the spatial and frequency dimensions.<sup>4</sup> The object wave is reproduced by a coherent optical response or a stimulated photon echo,<sup>5,6</sup> which occurs when a short read pulse is scattered from the recorded hologram.

It was shown<sup>7</sup> that, if a pair of correlated time-andspace-domain images are recorded in a hole-burning sample, then simultaneous readout of the two holograms causes interference between the coherent response amplitudes. By controlling the relative phase of the holograms one can use the photon-echo interference to erase selectively the data in the time domain<sup>8,9</sup> as well as to perform logical processing operations with light pulses.<sup>10</sup> It was also demonstrated that the interference between spectral holeburning holograms can be influenced by an external electric field by use of the Stark effect.<sup>11</sup>

In the past experimenters have controlled the interference by writing holograms with variable phase between the object and reference beams. For destructive interference the phase is changed by half the period, and for constructive interference the phase is constant. In this Letter we show that the interference can also be controlled by writing of timeand-space-domain holograms with different spectral envelopes while the phase between the object and reference beams has a constant value. Our considerations are based on the fact that, if during the writing of the hologram the reference pulse arrives earlier than the object pulse, then the stimulated echo occurs in the +1 diffraction order (i.e., in the direction of the object beam), whereas the amplitude in the -1 order is zero. The -1 diffraction order is canceled by causality because otherwise the echo would have to appear earlier than the read pulse. In the frequency domain causality implies that, whenever the hologram is recorded with a smooth spectral profile with a width larger than the inverse value of the time-domain path-length difference between the object and reference arms, the amplitude in the -1 order is zero.<sup>12</sup>

Consider two holograms with strongly modulated spectral envelopes,  $S_1(\nu)$  and  $S_2(\nu)$ , such that their sum,

$$S(\nu) = S_1(\nu) + S_2(\nu), \qquad (1)$$

is still a smooth spectral profile, as described above. The object and reference pulses have the same spectral envelopes, and they are overlapping in time. For either of the holograms the stimulated echo will then be emitted in both the +1 and the -1 orders. However, if the two holograms are combined, then their complementary envelopes will merge into one smooth spectral profile with a width larger than the inverse value of the time-domain path-length difference between the object and reference arms. Consequently the signal in the -1 direction is canceled, and one should observe destructive interference between the two holograms. On the other hand, the signal in the +1 diffraction order is not canceled, which means that constructive interference takes place.

In our experiment we use a 76-MHz repetition-rate picosecond dye laser synchronously pumped by a mode-locked frequency-doubled Nd:YAG laser (Coherent Antares 76). The spectral width of the dve-laser output is  $9 \text{ cm}^{-1}$ , which corresponds to an  $\sim$ 2-ps coherence time of the pulses. The dye-laser beam (Fig. 1) is directed through a Michelson interferometer with a fixed time delay  $\tau' = 20$  ps between the two arms and with nearly copropagating output beams. In this configuration the interferometer serves as a frequency filter with a cosinusoidal transmission function with a period  $(\tau')^{-1}$ . We obtain the complementary spectral functions by shifting the phase in one arm of the interferometer so that the frequency-domain fringe pattern moves by half the period (Fig. 2). In our case the spectral modulation means that each laser pulse is split into a pair of pulses so that the effective amplitude duration becomes equal to  $\tau'$ .



Fig. 1. Experimental setup: BE, beam expander; BS, beam splitter; M, spatial mask; H, hole-burning sample.



Fig. 2. Spectral envelopes at the output of the interferometer measured with a grating monochromator: (a), (b) Spectra used to record the first and the second holograms, respectively. The dashed curve is the spectrum of the picosecond laser.

The spectrally filtered laser beam is expanded to a diameter of ~1 cm and is then split into reference and object paths. The object beam passes through a mask and overlaps the reference beam at an angle of  $\alpha = 5^{\circ}$  at the sample, which is immersed in liquid He at T = 2 K inside an optical cryostat. A 120-mm focal-length lens (not shown) is placed between the mask and the sample and images the mask into the plane of the hologram. The object beam path is shorter by an amount  $\tau c$  that exceeds the ~1-mm coherence length of the laser pulses. On the other hand,  $|\tau| < |\tau'|$ , which means that the effective amplitudes of the object and reference beams overlap in time.

The hole-burning sample is similar to that described previously.<sup>2</sup> It consists of a 3-mm-thick block of polystyrene doped with octaethylporphin molecules at a concentration of  $10^{-3}$  mol/L. The absorption maximum of the  $S_1 \leftarrow S_0$  transition of octaethylporphin is at 618 nm and has an inhomogeneous width of several nanometers. The homogeneous spectral linewidth is of the order of 1 GHz.

We record the first hologram by illuminating the sample at an average intensity of  $1 \text{ mW/cm}^2$  and

an exposure time of 20 s with the spectrum shown in Fig. 2(a). We then switch to the complementary spectrum [Fig. 2(b)] and record the second hologram, using similar illumination conditions. Note that in both exposures the phase (and delay) between the reference and the object beams remains the same.

For the readout we illuminate the sample with the attenuated reference beam. The reconstructed image is detected with a CCD camera placed behind the cryostat in the direction of the object beam. Since in our case the delay of the object wave is negative, the light scattered in the direction of the CCD camera corresponds to the -1 diffraction order. By rotating the sample inside the cryostat by  $180^{\circ}$  around the vertical axis we can read out the hologram from the opposite direction, and the light diffracted toward the camera becomes equal to the +1 diffraction order.

The images recorded by the camera are shown in Fig. 3. The object masks used in the first and second exposures consist of a horizontal and a vertical bar, respectively, with their holographic images shown in Figs. 3(a) and 3(b). The response of the double-exposure hologram is presented below. Figure 3(c) shows the image observed in the -1 diffraction order displaying destructive interference, whereas in Fig. 3(d) the image observed in the +1 diffraction order exhibits constructive interference.

It is also possible to explain our present result in the time-domain picture in a way similar to that in Ref. 10. For this purpose we notice that in our experiment the interferometer and the beam splitter produce a series of four pulses altogether. Only one pair of pulses creates a hologram diffracting in the -1 diffraction order; the rest of the pulse pairs write holograms diffracting in the +1 order. Shifting the phase in the interferometer also produces a



Fig. 3. Images recorded by the CCD camera: (a), (b) Holographic image of the two different masks observed in the -1 diffraction order; (c) image from the double-exposure hologram observed in the -1 diffraction order, (d) the same double-exposure hologram observed in the +1 diffraction order.

phase shift between the pulses that write the hologram diffracting in the -1 order.

It should be noted that this description is straightforward only if the writing beams indeed consist of simple series of pulses with given time delays. On the other hand, with our considerations based on Eq. (1) there is no need to care about the temporal structure of the writing beams.

In the present experiment the efficiency of the photochemical hole-burning process is only a fraction of a percent, and therefore it takes seconds to record one time-and-space-domain hologram. By use of other types of spectrally selective material such as  $Pr^{3+}$ and  $Eu^{3+}$ -doped crystals, for which the efficiency and speed of the hole-burning process are much higher,<sup>13</sup> the recording time becomes much shorter. Since the complementary type of spectra can be generated simultaneously (e.g., by two corresponding interferometers) and there is no need to change quickly the optical path length between the object and the reference beam, conditions for practical applications can be achieved.

In conclusion, we have shown that, by superimposing two holograms with complementary spectral profiles in a spectrally selective persistent spectral hole-burning material, we can obtain simultaneously both destructive and constructive interference, which can be observed in different diffraction orders of the same double-exposure hologram.

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