Storage and time reversal of femtosecond light signals via persistent spectral hole burning holography

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Time and space domain holography by persistent spectral hole burning in photochemically active media is shown to permit storage, recall, and conjugation of temporal profiles of light signals as short as 100 fs. This limit for the temporal resolution is set by the finite spectral width of the impurity absorption bands of presently available recording materials.

Time and space domain holography in persistent spectral hole burning (PHB) media[1–4] has proven to be an attractive technique for ultrahigh speed optical data processing and storage.[5–7] The capability of this method to store and recall the temporal and spatial distribution of optical signals on a time scale of 2–3 ps has been demonstrated by Saari *et al.*[3] Time reversal,[2,3] polarization preserving storage,[8] as well as associative recall[9] of time-dependent signals with a few picosecond duration have been also accomplished.

The upper limit for the length ΔT of the pulsed signal is set by the phase relaxation time T_2 of the recording medium, corresponding to the minimum width of the spectral hole $\Delta\Omega > 1/\pi T_2$. A coherent superposition of the optical fields of different pulses over ΔT results and under certain conditions the temporal distribution is Fourier transformed into the spectral structure of the hole. In particular, the width $\Delta\omega_H$ of the hole envelope is the inverse of the pulse duration Δt.

The phase relaxation time amounts to $T_2 \approx 1$ ns which seems to be typical for purely electronic transitions in the impurity molecules of the recording medium at low temperatures.[10] Thus, the length of the optical pulse train is limited to about 100–200 pulses in the case of 3 ps time resolution. The corresponding transform-limited spectral bandwidth of 5–6 cm^{-1} ($\Delta\omega_L = 0.5/\Delta t$ for a Gaussian pulse shape) is 20–30 times narrower than the characteristic inhomogeneous absorption bandwidth $\Delta\omega_M$ of common hole burning media suggesting that the presently available hole burning media provide a potential to increase the time resolution to 0.1 ps. Such a time resolution would permit optical data processing at rates up to 10^{12}–10^{13} bits per second and increase simultaneously the length of the optical code to about 10^4 pulses. This simple up-scaling is based, however, on the unproven assumptions that (1) the hole burning efficiency is almost constant across the total extension of the absorption band, and that (2) nonlinear interactions between the different frequency components in the recording medium are negligible.

The exploration of these fundamental questions requires studies of the hole burning properties of PHB media with shorter optical pulses. In this letter we report on time domain holographic storage of optical pulse trains with pulse durations as short as 100 fs full width at half maximum (FWHM).

The optical pulses for the present investigation are generated in a colliding-pulse mode-locked (CPM) dye ring laser using the standard four-prism configuration for intracavity dispersion compensation.[11] This laser operating at a repetition rate of 110 MHz provides almost transform-limited pulses as short as 45 fs with a spectral bandwidth of 250 cm^{-1}, and of 60 mW average power for each of the two output beams. For the holographic studies the pulse duration is adjusted to about 70 fs. The laser wavelength is tuned to 621 nm by tuning the prism dispersion and adjusting the saturable concentration and the absorber jet position in order to achieve optimum overlap of the laser spectrum with the PHB absorption band of the samples. For the recording of the holograms we use samples which are similar to those of the earlier picosecond work[1–3] and which consist of 0.7-mm-thick polystyrene plates doped with molecules of octoethylporphine (OEP) or protoporphirine (PrP) at a concentration of 10^{-3}–10^{-4} mol/ℓ. During the experiments the samples are immersed in liquid-He pumped below the λ point. The maximum of the PHB active impurity absorption at low temperatures ($T \approx 1.8$ K) occurs at 619 and 622 nm for the OEP-doped and PrP-doped sample, respectively. The optical densities at these wavelengths are 1.0 for the OEP-doped and 0.5 for the PrP-doped sample. The FWHM of the absorption band amounts to 130 cm^{-1} for both types of samples.

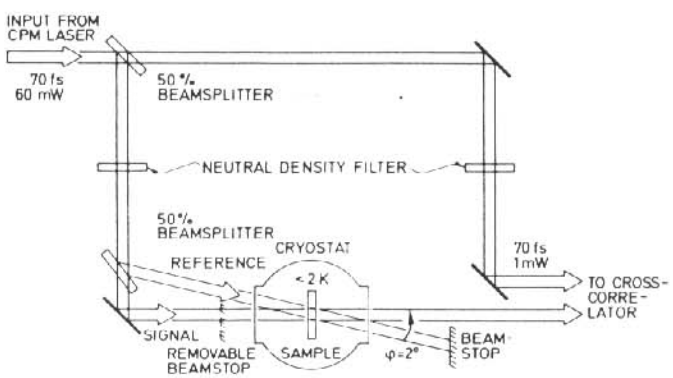

FIG. 1. Experimental setup.

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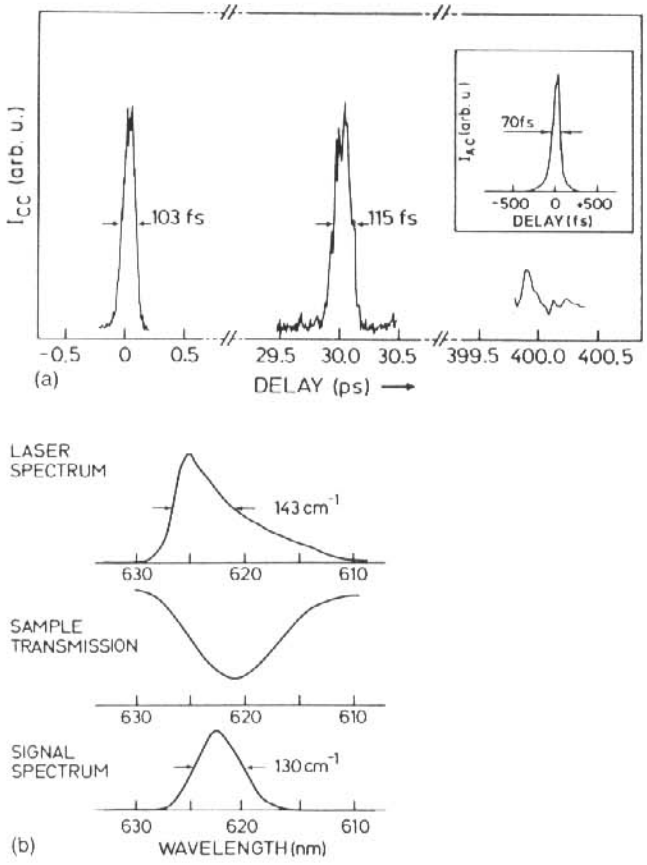

FIG. 2. (a) Cross-correlation traces of the read-out pulse (at zero delay) and recalled pulses (at 30 and 400 ps delays) with a short reference pulse. The insert shows the autocorrelation trace of the reference pulse. (b) Spectra of the laser pulse, PHB sample transmission, and the recalled signal.

A scheme of the experimental arrangement is presented in Fig. 1. Two unfocused laser beams derived from the same output of the CPM laser are superimposed on a 3.5-mm-diam spot on the sample. For the recording of the holograms the sample is illuminated with both beams. Coherent superposition of the temporally nonoverlapping optical fields belonging to the leading reference and the trailing signal pulse is provided by the long phase coherence time of the excited molecules in the storage medium.

Writing of the hologram requires an exposure level of 100 mJ/cm^2 corresponding to an exposure time of about 100 s for the CPM laser pulse energy of the order of 100 pJ or roughly 10^{10} writing pulse sequences. After the exposure the signal input is blocked and only the attenuated reference beam is passed through the hologram. In addition to the transmitted readout (reference) beam, a second weaker diffracted beam propagating into the direction of the original signal is observed which is the recalled signal.

Temporal analysis of the recalled signal is accomplished by measuring the cross-correlation profile using noncollinear sum frequency generation in a 0.7-mm-thick $LiIO_3$ crystal with an additional reference beam, split off from the CPM laser output

Figure 2(a) depicts cross-correlation traces corresponding to the signal recalled from a hologram in a PrP-doped sample with delays of 30 and 400 ps between the signal and reference pulses. The curve at zero time delay represents the cross correlation with the reading pulse. Compared to the original signal the image reproduced from the hologram exhibits about 10–20% of broadening. Assuming a sech^2 pulse shape, we obtain after deconvolution of the autocorrelation profile 75 fs duration of the recalled signal, instead of 66 fs for the original. Figure 2(b) shows the spectra of the recalled signal and the original together with the transmission of the sample in the region of the PHB active absorption band. The maximum of the recalled spectrum coincides with the absorption maximum and its width is equal to the 130 cm^{-1} spectral width of the impurity absorption band. This width is roughly 10% narrower than the spectral width of the original signal in accordance with the broadening observed in the time domain.

This result indicates that efficient hole burning is achieved over a broad inhomogeneous absorption band. Alongside with simultaneous hole burning no essential intermixing of spectral components takes place as is evidenced by negligible distortion of the recorded and recalled signals. Moreover, the present experiment achieves the ultimate temporal resolution, less than 100 fs, attainable from the applied storage medium, since the recalled spectrum is determined by the finite width of the PHB-active absorption band. Any further increase of the time resolution implies the availability of PHB systems with a broader inhomogeneous absorption bandwidth. Composite samples containing several overlapping absorption bands which may be realized by superposition of layers doped with different impurity species seem to be an attractive approach to overcome this limitation.

Figure 3 demonstrates the potential of PHB holography for storing and recalling the detailed temporal structure of a femtosecond pulse train which is produced by inserting a

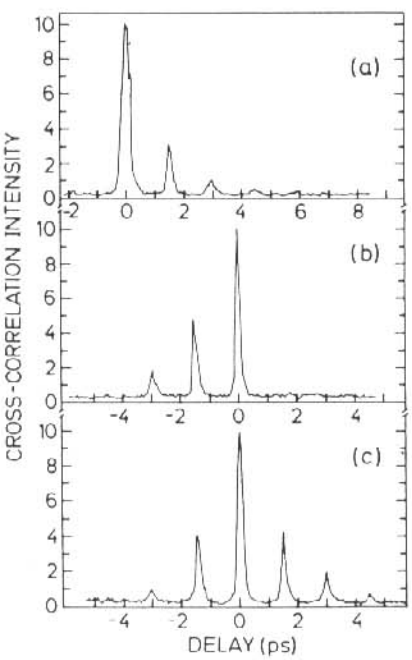

FIG. 3. Cross-correlation traces of the pulse trains, recalled from the holograms. (a) recorded with a leading reference pulse; (b) recorded with a trailing reference pulse; (c) recorded with a trailing reference pulse and read out with a pulse train.

0.25-mm-thick solid quartz plate Fabry–Perot étalon with $R = 65\%$ reflectivity optical coatings into the input signal beam. This signal is stored in a OEP-doped sample. The cross correlation of the recalled signal shown in Fig. 3(a) proves the excellent reproduction of the temporal profile of the initial signal pulse train.

Insertion of the Fabry–Perot étalon into the leading beam instead of the trailing beam during the burn-in exposure allows one to recall a time-conjugated signal. After storage of the hologram the étalon is removed from the leading beam which is used as a read-out pulse. A time-reversed replica of the original femtosecond pulse train [see Fig. 3(b)] is observed. Read-out of the hologram with the étalon still being present in the beam results in the formation of a time domain correlation profile of the read-out femtosecond pulse train with the stored pulse train [Fig. 3(c)].

In conclusion, our experiments have demonstrated the potential of the PHB-based holographic technique for time domain storage and processing of optical data on femtosecond time scales.

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