Time-resolved holography

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IN a conventional hologram, a photographic film records the interference pattern of monochromatic light, scattered from the object to be imaged, with a reference beam of unscattered light. Illumination of the developed film with a replica of the reference beam then creates a virtual image of the original object. Here we show how a molecular resonance can be used to record an interference pattern between light signals that arrive at different times, and with this technique create a hologram with time resolution. Using a timed reference pulse as a 'light shutter', we can record holographic images selectively, according to the time taken by light travelling from the object to the hologram. We use this method to image an object behind a semi-opaque screen, and indicate how a similar method could be used to inspect objects embedded in a dense scattering medium. Ultimately, this technique might be applied to the medical imaging of tumours.

Consider two optical pulses incident on a holographic recording medium as shown in the inset of Fig.1. Let pulse F arrive T_0 seconds before pulse G. If we use photographic film as the recording medium, then an interference pattern will be recorded only if the pulses overlap with each other at least partially in time. But suppose we replace the photographic film by a bank of resonators such as atoms, with each atom tuned to a slightly different optical frequency ω_j. In this case, the interference pattern of the two optical pulses F and G can be recorded even if the pulses are never present in the material at the same time[1].

If the optical pulse F is sufficiently brief, then its frequency spread will be wide enough to excite all of the atoms, much as a brief kick to a piano will excite all of the piano's strings. Each atom will continue to ring at its own natural frequency ω_j. After a delay time T_0, the second light pulse G arrives, and it will transfer energy either into or out of the jth atom depending on the relative phase between the optical electric field of G and the phase $\phi_j = \omega_j T_0$ of the still-ringing atom. Because each atom's phase depends on its particular resonant frequency ω_j, a given time delay T_0 between the two incident light pulses will produce a unique pattern of excited and unexcited atoms in frequency space.

After interacting with both of the light pulses F and G, let the absorption of the jth atom be permanently altered by an amount proportional to the atom's final energy (by a mechanism described below). If the now altered atoms are illuminated by a replica of pulse F, they will absorb and re-radiate light, again at the frequencies ω_j appropriate to each atom. At first, the phases of the re-radiated light will be incoherent, but after a time of exactly T_0, the phases of the different frequencies come together to reproduce coherently a duplicate of the pulse G (ref. 1). It can be shown[2], however, that application of a replica of pulse G to the altered atoms causes a re-radiation of light that loses rather than gains phase coherence with the passage of time. Unlike conventional off-axis holography, where either light beam can be used to reconstruct the other, the bank of atoms here records the direction of time's arrow.

In fact, as we describe below, it is possible to reproduce an image of the scattered light that strikes the hologram before the reference pulse arrives. To produce such an image, the hologram is read out by a replica of pulse F applied in the opposite direction to the original pulse: reversing the spatial orientation of the readout pulse reverses the relative phase relation of the re-radiated light, and thereby leads to the recreation of the 'before' rather than 'after' scattered light.

Now consider illuminating an entire scene with a pulsed laser. Record the reflected light G using a bank of tuned resonators, and let a reference pulse F also illuminate the resonators, as described above. If the reference pulse arrives at the resonators before any of the reflected light, then the entire scene can be recalled by simply reading with another reference pulse. But if the initial reference pulse is delayed so that it arrives after some of the reflected light, then reading (with another reference pulse) will only recreate the parts of the image that arrived after that first reference pulse, which are the parts of the scene that were located farthest from the resonators. For example, if the scene consisted of the street view of a bookshop, then, with a suitably timed reference pulse, the reconstructed image would show the books on display deep inside the store, and would not show the reflections off the shop's front window.

We constructed a bank of narrow-band resonators by doping a solid block of polystyrene with the organic dye protoporphyrin[2-6]. Each dye molecule acts as a lightly damped resonator, but because of inhomogeneities in the plastic matrix, each dye molecule has a slightly different resonant frequency. When illuminated by narrow-band light of frequency ω, the molecules that happen to be in resonance with the light become excited. A fraction of these excited molecules subsequently relaxes into a metastable state (thought to be a tautomerization of the original molecule). Once in this transformed state the molecule's absorption is shifted to a completely different spectral region, so a narrow 'hole' is burned into the sample's absorption spectrum at the frequency ω. This spectral hole remains as long as the sample is kept cold. If cooled to a temperature of 2 K, the phase-relaxation time, T_2, of the molecule's upper level becomes quite long ($T_2 \approx 1$ ns), so that the absorption hole has a narrow homogeneous spectral width of $\Delta\omega = 1/(\pi T_2) \approx 0.01\ \mathrm{cm}^{-1}$. The polystyrene matrix causes the net absorption spectrum of all of the molecules to form an inhomogeneously broadened band extending over a range of $200\ \mathrm{cm}^{-1}$. Consequently, this material resembles a bank of $200/0.01 = 20{,}000$ narrow-bands of resonators and we use this 'spectral-hole-burning' material to record and store the spectral (and, as we show below, spatial) contents of an incident light beam.

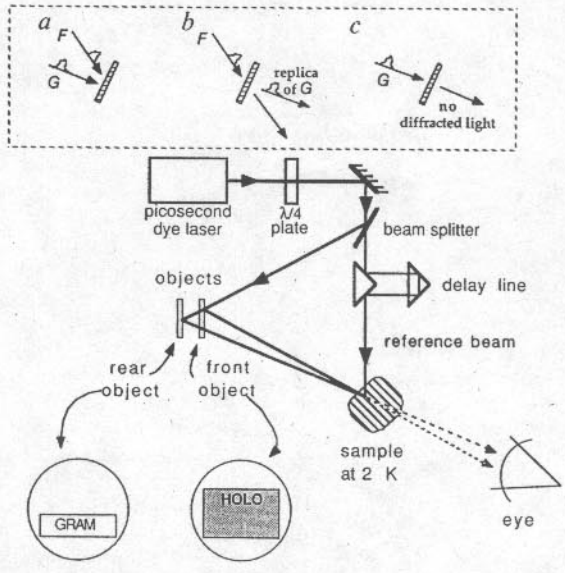

FIG. 1 Main figure: experimental set up. The front object is a frosted slide with the letters 'HOLO' pasted on the front. The rear object is the letters 'GRAM' pasted on the back of the same slide. Inset: *a*, Writing the hologram with two light pulses separated in time. *b*, Reading with the earlier pulse F recreates the later pulse G. *c*, Reading with the later pulse G does not produce any diffracted pulse.

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In our experiments, we used a 3-mm-thick block of polystyrene doped with protoporphyrin at a concentration of 10^{-3} mol l^{-1}. The useful sample area was 4 cm^2. The sample was immersed in liquid helium, and the helium vapours were pumped to reduce the temperature to 2 K. The peak of the broad absorption feature was at $\lambda = 621$ nm, at which the optical density was 1.6. The light source was a continuous-wave modelocked Nd:YAG laser (Coherent Antares 76s) which synchronously pumped a tunable dye laser (Coherent 701) to produce pulses having an intensity width of 8 ps full width at half maximum (FWHM). (These pulses are not transform-limited; they have a coherence width of only 0.5 ps.) The repetition rate of the laser pulses was 76 MHz. A beamsplitter divided the beam from the picosecond dye laser into a reference beam and a separate beam to illuminate the various objects in the scene, as shown in Fig. 1. The reference beam was expanded by a telescope to illuminate the entire polystyrene block. Scattered light from the illuminated objects simply propagated to the polystyrene block with no intervening lens. The angle between the reference beam and the image-bearing beam was ~14°.

The recorded scene consisted of two objects. The nearby object was a 1.0-mm-thick glass slide with the letters 'HOLO' attached to its front surface. The distant object was a white paper screen carrying the letters 'GRAM', which was pressed against the back of the transparent slide, so the separation between the two objects was ~1 mm. To increase the amount of light scattered by the slide, its front surface was coated with a frosting aerosol spray (New York Bronze Powder Co.) The slide was illuminated from the front. Of the scattered light from the slide reaching the polystyrene block, ~80% came from the front sprayed surface of the slide, and only ~20% came from the rear surface. Viewing the laser-illuminated slide by eye from the position of the polystyrene block, one could clearly read the words HOLO, but the intense glare from the front surface almost completely obscured the letters GRAM located near the back of the slide.

Light from the laser reached the front of the slide 5 ps before it reached the back of the slide. Consequently, light from the front of the slide reached the storage medium 10 ps (twice the glass travel-through time) before light from the back of the slide. In the first experiment a hologram was recorded with the reference beam timed to arrive before both of these object waves. In the second experiment a hologram was recorded with the reference beam carefully timed to arrive within the 10-ps interval between the two object waves.

The reference beam and the object beam each had an average intensity of 0.2 mW cm^{-2} at the location of the storage medium. An exposure time of 2–3 minutes was needed to record a hologram with a fluence of 70–100 mJ cm^{-2}, corresponding to 10^{10} identical pairs of laser pulses. To write the first hologram we tuned the laser wavelength to the absorption maximum of the medium at 621 nm (19,950 cm^{-1}). Because the absorption spectrum is 200 cm^{-1} wide and the dye-laser pulses have a spectral bandwidth of only ~30 cm^{-1}, we could change the wavelength of the dye laser by ~30 cm^{-1} and then record a new hologram in a 'fresh' spectral region of the storage medium without affecting any previously stored holograms. The different holograms written at different centre wavelengths were later selectively read out by simply adjusting the centre wavelength of the reading dye-laser pulse. In the absence of all illumination these holograms lasted for as long as the sample was kept cold.

We read the holograms by blocking the light coming from the objects and illuminating the hologram with only the reference beam. Figure 2*a* shows the reconstructed image when the

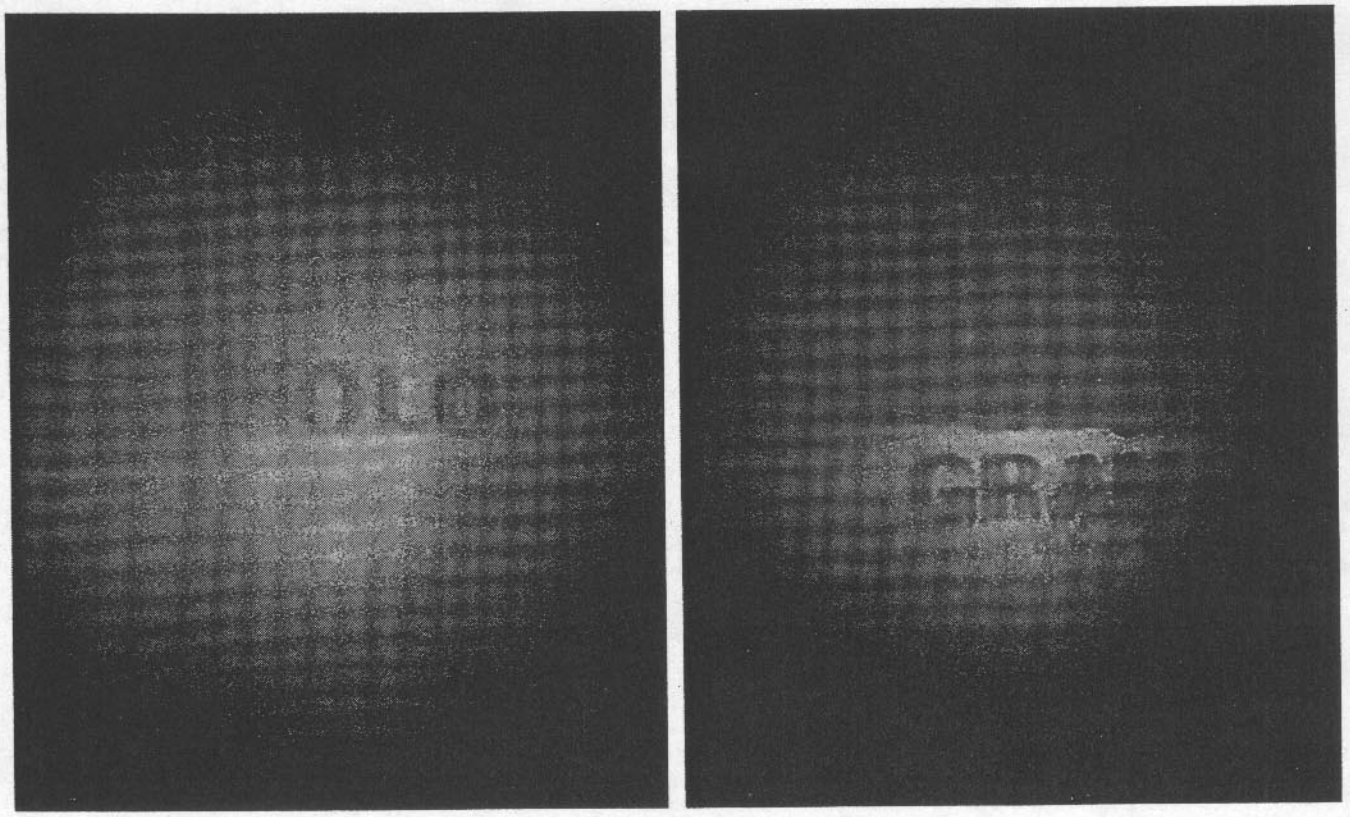

FIG. 2 *a*, Holographic reconstruction of both objects. The reference pulse was set to arrive before light from either of the objects. The glare from the frosted glass in front with the word HOLO obscures the object GRAM behind it. *b*, In this hologram the reference pulse was delayed to arrive after light from the front object (frosted glass) but before light from the rear object. The front object is no longer reconstructed, whereas the rear object (GRAM) is now plainly visible.

hologram was recorded using a reference beam that arrived a few picoseconds before any of the light from the glass slide. The letters HOLO on the front of the slide are plainly visible, but the glare from the front of the slide almost obscures the letters GRAM near the back of the slide. Figure 2*b* shows the reconstructed image when the reference beam pulse was carefully set to arrive after light from the distant object but before light from the nearby object. Now only the distant object was reconstructed, and the nearby object was eliminated. We emphasize that the light from these objects need not arrive at the polystyrene block at the same time as light in the reference pulse for the hologram to be recorded. In fact, because the coherence time of the light pulses was only 0.5 ps, and because we set the reference beam to arrive at the storage medium ~ 5 ps before the light from the back of the slide, the reference and object beams could not have produced a conventional intensity interference pattern in the storage medium. We note that the maximum time delay permitted between the object and reference beams was 10^3 ps and is set by the phase-decay time T_2 of the sample.

In the experiment reported here we could selectively reconstruct those objects that sent light to the hologram after the reference pulse had arrived. For some applications, however, it is desirable to do the opposite and recreate the light that arrived before the reference pulse. For example, consider the problem of imaging an object that is embedded in a scattering medium, such as a tumour embedded in breast tissue. Illuminate the tissue from behind with a short laser pulse. Light transmitted through the tissue without any scattering will emerge before light that has been multiply scattered by the tissue[7]. Because the eye records all of this light, and because the scattered light overwhelms the unscattered light, the tumour remains unseen. But if light that arrived earlier at the eye could be selectively enhanced, than a shadowgram of the tumour would become visible.

This selection of early light over late light can be accomplished by simply altering the direction of the readout beam used in the experiments above. Instead of using a readout beam in the same direction as the reference beam, one should use a readout beam that is directed exactly opposite to the direction of the original reference beam. This is the 'four-wave mixing' geometry of traditional phase-conjugation experiments[8], but with a spectral-hole-burning material now only the light that arrived before the reference beam is holographically reconstructed in the final image. In this way light scattered from back-lighted tissue could be eliminated, while light travelling directly through the tissue could be preserved, and would form a shadowgram of features embedded in the tissue. We caution that this scheme requires a very large dynamic range for the hologram, because in thick tissue the scattered light can be much more intense than the unscattered light. We estimate that our present spectral-hole-burning medium has a dynamic range limited to $\sim 10^3$. Experiments to demonstrate such selective imaging are in progress. □

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