Volume holographic pulse shaping

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Volume holograms for subpicosecond pulse shaping are described. Experimental results with a camphorquinone-doped plastic hologram probed by a colliding-pulse mode-locked laser are presented. We observe a 2-THz beat frequency in the diffracted pulse.

The use of spectral filtering to modulate subpicosecond laser pulses has been detailed in recent research.1-4 We describe experiments that achieve similar results by using the Bragg selectivity of volume holograms to control spectral channels independently. Volume holographic techniques offer high spectral resolution and the potential for high-speed programming. In addition to applications in high-bandwidth communications and nonlinear molecular spectroscopy, these techniques are important for constructing programmable pulses for polychromatic volume holographic recording systems.5 Subpicosecond effects in photorefractive volume holograms were previously observed by Acioli et al.,6 who attribute their results to spatial localization of holographic regions rather than to the spectral filtering process reported here.

Our goal is to diffract an incident plane-wave pulse onto a diffracted plane-wave pulse with a modulated temporal spectrum. Just as with thin holograms, a single-grating volume hologram diffracts different wavelengths in different directions. For a volume hologram, however, the diffraction efficiency varies strongly with wavelength. In the limit as the hologram becomes infinitely thick, only a single wavelength is diffracted from a pulse incident from a given direction. Suppose that the nominal wave vector of the incident pulse is \( \mathbf{k}_1 \). We wish to diffract onto an output pulse with nominal wave vector \( \mathbf{k}_2 \). Figure 1 shows these vectors on the wave normal surface for light of frequency \( \omega_0 \). \( \theta_1 \) and \( \theta_2 \) represent the angles between \( \mathbf{k}_1 \) and \( \mathbf{k}_2 \), and the \( \hat{z} \) axis. Ideally, \( \theta_1 = \theta_2 = \theta \). Also shown are the wave normal surfaces for frequencies clustered around \( \omega_0 \). The incident pulse consists of a superposition of plane waves with wave vectors parallel to \( \mathbf{k}_1 \). We would like the diffracted pulse to be a similar superposition of plane waves propagating along the \( \mathbf{k}_2 \) direction. The Bragg phase-matching constraint tells us that a volume grating couples a pair of plane waves if and only if the grating wave vector joins the end points of the wave vectors of the plane waves on the wave normal surface. The grating wave vectors shown in Fig. 1 therefore couple the plane waves that they join. The grating wave vector that couples light at frequency \( \omega \) is

\[
K_\omega(\omega) = 2 - \frac{\omega}{\omega_0} |\mathbf{k}_1| \sin \theta \hat{\mathbf{\hat{x}}}. \tag{1}
\]

As shown in Fig. 1, all the grating wave vectors that couple one pulse to the other are parallel. For low-diffraction-efficiency holograms, the phase and amplitude of the diffracted signal at frequency \( \omega \) is proportional to the phase and amplitude of the grating at spatial frequency \( K_\omega(\omega) \). This qualitative analysis suggests that a hologram consisting of many parallel gratings is appropriate for the pulse-shaping problem. We limit our analysis to such one-dimensional holograms.

We focus on the system shown in Fig. 2. The subsystem consisting of a cw Ar+ laser and acousto-optic (AO) deflectors is used to record a volume hologram. Once the hologram has been recorded, it is probed by a pulse from a mode-locked source. Autocorrelations of the diffracted and undiffracted pulses are measured. During the recording process, temporal signals \( s_1(t) \) and \( s_2(t) \) are fed into the AO cells. The spatial spectra of the signals diffracted by the AO cells correspond to the Fourier transforms of \( s_1(t) \) and \( s_2(t) \). The symmetric arrangement of the AO cells guarantees that the recorded hologram varies only along the \( \hat{x} \) axis because the linear variation of the Doppler shift of the AO cell signals with diffraction angle means only beams whose wave vectors have the same \( \hat{z} \) component yield interference patterns with nonzero time-average amplitude. No cross gratings form. The magnitude of the recorded grating at spatial frequency \( K_\omega \) is proportional to the product of the magnitudes of the spectra of \( s_1 \) and \( s_2 \) at frequency

\[
\Omega = \frac{2\pi v_A}{\lambda} \left( \tan \theta - \frac{K_\omega}{4\pi \cos 2\theta} \right), \tag{2}
\]

where \( v_A \) is the acoustic velocity and \( \lambda \) is the wavelength of the recording field. The phase of the grating is equal to the difference in the phases of the spectra at \( \Omega \). The mode-locked pulse scatters off a volume hologram rather than directly off the AO cells because holograms support greater thickness and resolution than are practical with AO cells. Despite these difficulties direct AO modulation may play a role in future applications.
Once recorded, the hologram is a perturbation to the permittivity of the volume. The perturbation gives rise to a scattered beam that is a filtered version of the incident pulse. We find an expression for the scattered field using the wave equation
\[ \nabla \times \nabla \times E - \varepsilon(x) \partial^2 E / \partial t^2 = 0, \] (3)
where \( \varepsilon(x) \) is the holographic perturbation. For TE modes, Eq. (3) reduces to the scalar integral equation
\[ \Psi(x, z, \omega) = \Psi_0(x, z, \omega) - j \omega^2 \mu \varepsilon \int \int H^{(0)}_0(\omega \sqrt{\mu \varepsilon} |\rho - \rho'|) V(x') \Psi_0(\rho', \omega) d\rho' d\rho, \] (4)
where \( H^{(0)}_0 \) is a zeroth-order Hankel function, \( \rho = x^2 + z^2 \), \( \Psi_0 \) is a solution to the unperturbed wave equation, and we have taken a temporal Fourier transform. As we wish to maintain a linear relationship between the spatial spectra of the recording beams and the spectrum of the scattered beam, we consider Eq. (4) in the weak scattering limit, where the Born approximation is appropriate. We decompose \( \Psi \) into the sum of an incident part \( \Psi_0(x, z, \omega) \) and a scattered part \( \Psi_s(x, z, \omega) \). The Born approximation consists of the assumption that \( |\Psi_s| \ll |\Psi_0| \) for all \( \rho, \omega \). We substitute \( \Psi_0 \) for \( \Psi \) and \( \Psi_s \) on the right-hand side of Eq. (4) to find the scattered spectrum,
\[ \Psi_s(\rho, \omega) = -j \omega^2 \mu \varepsilon \int \int H^{(0)}_0(\omega \sqrt{\mu \varepsilon} |\rho - \rho'|) V(x') \Psi_0(\rho', \omega) d\rho' d\rho. \] (5)
In the Fraunhofer regime, where \( \rho = |\rho| \gg L_x, L_z, \)
\[ H_v^{(0)}(\omega \sqrt{\mu \varepsilon} |\rho - \rho'|) \]
\[ = \sqrt{2c / \pi \omega \varepsilon} \exp(j \pi / 4) \exp(j \omega \mu \varepsilon (\rho - \rho')), \] (6)
where \( \rho = k_0(\rho / \rho), k_0 = \omega \sqrt{\mu \varepsilon} = 2\pi / \lambda \), and \( \epsilon \) is the speed of light in the hologram. The incident pulse is a solution to the unperturbed wave equation,
\[ \Psi_0(\rho, t) = f(t - k_0 \cdot \rho) \exp[j(\omega t - k_0 \cdot \rho)], \] (7)
where \( |k_0|^2 = k_0^2 \). The Fourier transform of the incident pulse is
\[ \Psi_v(\rho, \omega) = \exp(j \omega k_0 \cdot \rho) F(\omega - \omega_0), \] (8)
where \( F \) is the Fourier transform of \( f \). Substituting in Eq. (5), we find that
\[ \Psi_s(\rho, \omega) = \exp(j \omega k_0 \cdot \rho) F(\omega - \omega_0) T(\omega, k_0), \] (9)
where
\[ T(\omega, k_0) = \exp(-j \pi / 4) \omega^2 \mu \varepsilon \sqrt{2c / \pi \omega \varepsilon} \int \int \exp[j(\omega k_0 \cdot (k_0 - k_0) - \rho)] V(x') d\rho' d\rho. \] (10)
As we saw in our qualitative discussion, the transfer function at \( \omega \) is proportional to the Fourier transform of \( V(x) \) at \( K_g = (\omega / \omega_0)(k_0 - k_0) \).

To explore the transfer function experimentally, we use discrete gratings. When a single grating of spatial frequency \( K_g \) is recorded, \( V(x) = \Theta[V_0 \exp(-j K_g x)] \). Suppose \( k_0 \) makes an angle of \( \theta = 0 \) with respect to the \( z \) axis and \( k_0 \) makes an angle \( \theta = \theta + \Delta \theta \), where \( \Delta \theta \ll 1 \). Equation (10) becomes
\[ T(\omega, \Delta \theta) = \exp(-j \pi / 4) \omega_0 \varepsilon \mu \varepsilon \sqrt{2c / \pi \omega_0 \varepsilon} \sin(2k_0 / \omega_0 \varepsilon \cos \theta + 2k_0 / \omega_0 \varepsilon \sin \theta - K_g L_z / 2) \]
\[ \times \exp\left(\frac{k_0 \omega_0 \Delta \theta \sin \theta - K_g L_z / 2}{2} \right) \]
\[ \times \frac{k_0 \omega_0 \Delta \theta \sin \theta}{k_0 \Delta \theta \sin \theta}, \] (11)
where we assume that the perturbation is nonzero for \( -L_x / 2 < x < L_x / 2 \) and \( -L_z / 2 < z < L_z / 2 \), and we limit ourselves to transmission holograms. If we assume that \( L_x \cos \theta > L_z \sin \theta \), the transfer function is nonzero over a range of \( \Delta \theta \) approximated by
\[ |\Delta \theta| \leq \frac{\lambda_0}{L_z \sin \theta}. \] (12)
Although the angular spread of the diffracted pulse allows for further pulse shaping by spatial filtering, in our experiments the autocorrelation was generated from the total diffracted field, which corre-
Fig. 3. (a) Autocorrelation of the undiffracted pulse passing through a two-grating hologram. (b) Autocorrelation of the corresponding diffracted pulse.

responded to an integral of the autocorrelation signal over $\Delta \theta$. The bandwidth of this signal is determined by finding the range of frequencies $\Delta \omega$ for which $T$ is nonzero over the range of the allowed $\Delta \theta$. The center frequency at a given $\Delta \theta$ is

$$\omega_B(\Delta \theta) = \frac{cK_g}{\Delta \theta \cos \theta + 2 \sin \theta},$$

and the range of frequencies for which diffraction occurs is

$$\Delta \omega = \left(1 + \frac{2}{K_gL_x}\right)\omega_B\left(\frac{2}{h\omega_L \sin \theta}\right) - \left(1 - \frac{2}{K_gL_x}\right)\omega_B\left(-\frac{2}{h\omega_L \sin \theta}\right) \approx \frac{2\pi c}{\tan \theta \left(\frac{1}{L_x \sin \theta} + \frac{1}{L_x \cos \theta}\right)}.$$ \hspace{1cm} (14)

In our experiments, $L_x = L_x = 2\text{ mm}$ and $\theta = 22^\circ$. In this case $\Delta \omega$ corresponds to a spectral width of 2 nm on a center wavelength of 620 nm. For weak scattering, the transfer function for multiple gratings is the superposition of the individual transfer functions. For two gratings corresponding to wave vectors $K_{g1}$ and $K_{g2}$, one should observe the beat frequency,

$$\Omega \approx \frac{c(K_{g1} - K_{g2})}{2 \sin \theta}.$$ \hspace{1cm} (15)

We used holographic material created by doping 5% camphorquinone (TCI America) into a commercial casting plastic (Castolite-AP) as a holographic medium. The sample is cast between glass slides. The sample used in the experiment described here is 2 mm thick. The holographic mechanism in this material is a hydrogen-ion transfer with the polymer matrix. In our samples the hologram is permanent. The holograms were recorded using the 514-nm line of an Ar$^+$ laser. The diffraction efficiency of the recorded holograms was 1%. The weak diffraction efficiency made the dye amplifier chain of Fig. 2 necessary. The pulse repetition frequency from the chain was 10 Hz. We recorded a two-grating hologram with a nominal grating period of 686 nm. The separation between the periods of the two gratings is 6 nm, corresponding to a frequency difference of 14 MHz for the two harmonics in the AO cells. This separation corresponds to a beat frequency of 1.9 THz in the diffracted pulse.

We reconstruct the hologram using a 620-nm pulse. Figure 3 shows the autocorrelations of the undiffracted and diffracted pulses passing through this hologram. Asymmetry in the autocorrelation arises from the long integration time (~30 min) used to take the curve. This integration time was needed to overcome the low repetition frequency of the amplifier chain. The ~250-fs peak spacing in the diffracted pulse autocorrelation is consistent with a 2-THz beat frequency.

While we have chosen to work in the weak diffraction limit, high-diffraction-efficiency volume holograms may be needed for the technique described here to be competitive with other methods. It may also be desirable to consider two-dimensional perturbations to incorporate dispersion compensation, combat spatial dispersion in the diffracted spectrum, and improve the spectral resolution. While permanent holograms have been used for this demonstration, the use of dynamic media is essential to the success of this technique. Photorefractive and dynamic organic media can be used to demonstrate pulse shaping with millisecond programming times. Extension to semiconductor media may allow pulse shaping with nanosecond programming times. Short programming times will likely require a shift to volume holography in slab waveguides, where many grating holograms can be recorded with higher diffraction efficiency than is possible in bulk and where resonant nonlinearities may be used to record relatively fast and sensitive dynamic gratings.

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References