Control of Light Transmission through Opaque Scattering Media in Space and Time

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We report the first experimental demonstration of combined spatial and temporal control of light transmission through opaque media. This control is achieved by solely manipulating spatial degrees of freedom of the incident wave front. As an application, we demonstrate that the present approach is capable of forming bandwidth-limited ultrashort pulses from the otherwise randomly transmitted light with a controllable interaction time of the pulses with the medium. Our approach provides a new tool for fundamental studies of light propagation in complex media and has the potential for applications for coherent control, sensing and imaging in nano- and biophotonics.

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Concentrating light in time and space is critical for many applications of laser light. Broadband mode-locked lasers provide the required ultrashort light pulses for multiphoton imaging [1,2], nanosurgery [3], microstructuring [4], ultrafast spectroscopy [5,6], and coherent control of molecular dynamics or of nanooptical fields [7–9]. Multiple random scattering in complex media severely limits the performance of these methods, but often is an unavoidable nuisance in many systems of interest, such as biological tissue or nanophotonic structures [10]. Spatially, random scattering strongly distorts a propagating wave front, creating the well-known speckle interference pattern [11]. In the time domain, ultrashort pulses are strongly distorted and widely stretched due to the broad path length distribution in multiple scattering media [12]. These temporal and spatial distortions are not separable [13].

There is a strong interest in improving applications of ultrashort laser pulses in complex scattering media. Phase conjugation has been applied to spatially focus light from a short-pulse laser source through a thin scattering layer [14]. Similarly, phase conjugation is applied to correct distortions of the ballistic wave front to improve the resolution of two-photon microscopy [15]. Coherent control of two-photon excitation through scattering biological tissue has been demonstrated [16]. Those experiments share the common limitation that the control is limited only to those photons that take the shortest paths through the disordered media and arrive at the target volume without being multiply scattered.

Recently it was demonstrated that random scattering can actually be beneficial rather than detrimental for the performance of optical systems. Applying a shaped wave front of monochromatic light to a strongly scattering medium, Vellekoop *et al.* achieved spatially controlled focusing in transmission [17] and on fluorescent molecules inside the medium [18]. These findings have opened new possibilities for imaging in optically thick biological matter [19] and allow trapping particles through turbid media [20]. All of these studies used monochromatic light sources, and therefore only allowed spatial control over the scattered light. Related techniques which allow coherent focusing in scattering media are known from ultrasound [21] and microwaves [22]. The frequency of those types of waves is low enough that electronic transducers can be used to time reverse waves, which redirects the waves towards their source. This technique has successfully helped to improve imaging resolution [23] and communication bandwidth [24].

In this Letter we generalize the concept of wave front shaping to the regime of broadband light. We report the first experimental demonstration of combined spatial and temporal control of light transmission through random scattering media. By only controlling spatial degrees on freedom of the incident wave, we control the field amplitude at a selected point in space and time behind the sample. This enables us to create an ultrashort pulse from the otherwise randomly transmitted light. We can control the amount of time the optimized pulse stays in the sample and thereby select the path length of the light through the medium.

In Fig. 1 we show a simplified scheme of our experimental realization. Pulses from a Ti:sapphire laser (duration 64 fs, center wavelength 795 nm) illuminate a two-dimensional phase-only spatial light modulator (SLM). The SLM pixels are grouped into N independent segments each of which induces a controllable phase shift $\Delta\Phi_i$. The light is subsequently focused onto a layer of strongly scattering titanium dioxide pigment (thickness $L = 13.5\ \mu$m, transport mean free path: $l_t = 1\ \mu$m [25]). The transmitted light appears as a spatiotemporal speckle pattern. In the experiment, we optimize the field amplitude at a selected point in space and time. A pinhole fixes the

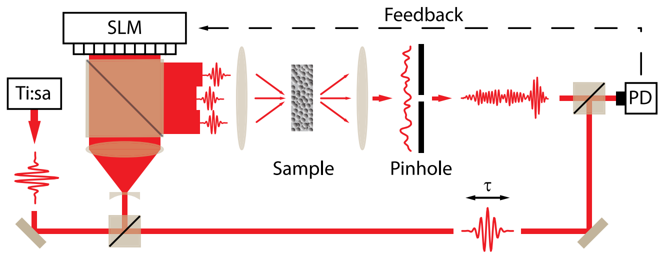

FIG. 1 (color online). Experimental setup (see text).

spatial coordinate. To fix the time, the speckle pulse is overlapped with a reference pulse in a heterodyne detection scheme in a configuration similar to [26]. The heterodyne signal exactly corresponds to the cross correlation of the speckle pulse and the reference pulse [27]. Effectively, it is an instantaneous measurement of the transmitted field amplitude at the delay time of the reference pulse τ. This signal serves as feedback for an optimization algorithm, which controls the incident wave front via the SLM.

The principle of the experiment can be described as follows. Light reflected from a single segment on the SLM is transmitted through the sample, giving rise to the field $E_i(t)$ at the detector. Its phase can be modified by a time-independent phase shift $\Delta\Phi_i$ via the SLM. The total field scattered into the detector $E_{\rm out}(t)$ is therefore given by the sum over all segments

$$E_{\rm out}(t) = \sum_{i=1}^{N} E_i(t) e^{i\Delta\Phi_i}. \quad (1)$$

Multiple scattering allows us to assume that the contributions $E_i(t)$ from the different segments at every single point in time t are uncorrelated random variables with Rayleigh distributed amplitudes $|E_i(t)|$ and uniformly distributed phases $\Phi_i(t)$ [28]. For the nonoptimized case, the resulting field $E_{\rm out}(t)$ can be viewed as the result of a random walk in the complex field plane. After the optimization, all contributions are in phase, adding up constructively. The average amplitude enhancement is given by [17]

$$\langle\alpha\rangle = \frac{\langle|E_{\rm opt}|\rangle_{\rm rms}}{\langle|E_{\rm rnd}|\rangle_{\rm rms}} = \left(\frac{\pi}{4}(N-1)+1\right)^{1/2} \approx \left(\frac{\pi}{4}N\right)^{1/2}. \quad (2)$$

Since the instantaneous field amplitude is optimized, the instantaneous intensity accordingly increases, with an average intensity enhancement $\eta = \alpha^2$. The simple model leading to Eq. (2) does not provide a prediction for the resulting pulse duration. We address this point later in this Letter.

The nonoptimized data were obtained by setting random phase values to the SLM segments. The optimization algorithm adjusts the phase shifts $\Delta\Phi_i$ such that the amplitude of the heterodyne signal is maximized. We performed the optimization at 20 equidistant time delays $\tau_{\rm opt}$ between -1.05 ps to $+13.6$ ps. For each $\tau_{\rm opt}$, the optimization was performed four times, with $N = 12$, 48, 192, and 300 segments, respectively, each time starting from a new random phase pattern.

Our main result is displayed in Fig. 2, showing the amplitudes of both the nonoptimized and the optimized pulses for different time delays $\tau_{\rm opt}$ and $N = 300$ segments on the SLM. The long time-tail of the average nonoptimized transmission reflects the broad path length distribution which has been observed in similar earlier studies [12]. The optimized amplitudes show sharp, distinct peaks with dramatically increased amplitudes at the desired time delay. We can control the amount of time the optimized pulses stay in the sample by the time delay $\tau_{\rm opt}$, and by that we control the path length of the pulses through the sample. Note that the heterodyne signal is proportional to the field amplitude, the intensities exhibit even more pronounced optimized peaks.

The enhancement α versus time delay $\tau_{\rm opt}$ is shown in Fig. 3. Its magnitude, depending on the number of segments on the SLM, is constant from zero to several picoseconds time delay. This result shows that our method works for short light paths as well as for light paths more than 10 times longer than the sample thickness.

For long time delays a continuous decrease of α is observed, which is related to the noise level of the experiment. We include a quantitative analysis of this effect in the supplemental material [27].

Figure 4 shows the average enhancement in the constant regime in Fig. 3 versus N together with the enhancement expected from theory [Eq. (2)], $\langle\alpha\rangle = \sigma(\frac{\pi}{4}N)^{1/2}$. The prefactor $\sigma = 0.90$ corrects for the nonuniform illumination

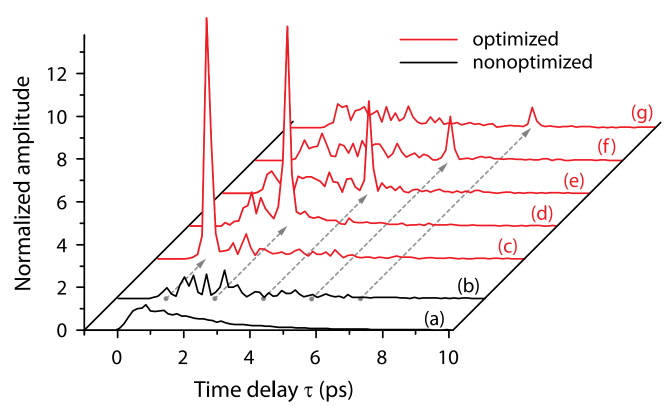

FIG. 2 (color online). Optimized and random speckle pulses. (a) Amplitude of heterodyne signal of a nonoptimized pulse as a function of time delay, averaged over 50 random speckle pulses. (b) Typical single random speckle pulse. (c)–(g) Amplitudes of single pulses after optimization at different time delays which are indicated by the dashed arrows. The optimization has been performed by dividing the SLM into 300 segments. The optimization generates strong, short pulses from diffuse light. The zero delay position is at the maximum amplitude with no sample. The plotted curves have been normalized to the maximum of the average nonoptimized heterodyne signal (factor 1.53 $\rm mV^{-1}$).

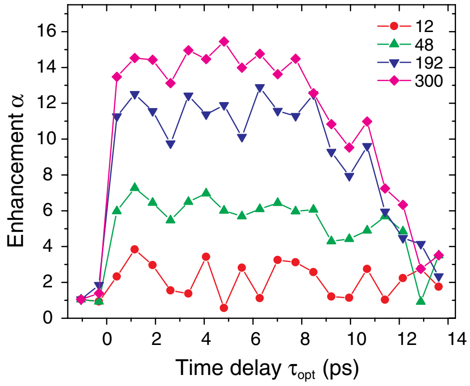

FIG. 3 (color online). Enhancement α versus selected time delay τ_{opt} for different number of segments N on the spatial light modulator.

of the SLM surface with a truncated Gaussian beam, which effectively leads to a reduction of the number of used segments [27]. Our model matches the data well with no adjustable parameters.

The measurement of the output pulse duration reveals an interesting characteristic the optimization method. Independent of the time delay, the optimized pulses have an (intensity) duration of $\Delta t_{\mathrm{opt}} = 115$ fs, calculated from the average width of the cross-correlation peaks (Fig. 5). The optimized pulses are lengthened compared to the input pulses ($\Delta t_{\mathrm{in}} = 64$ fs). What follows is a qualitative explanation of this effect. Further details are included in the supplemental material [27]. The increased duration corresponds to spectral narrowing due to the optimization procedure. Transmission through the random medium introduces strong random phase and amplitude fluctuations within the laser bandwidth (frequency speckle) [28]. This means that a single segment of the SLM, which adds an almost frequency-independent phase shift, cannot optimize all frequencies equally well. The optimization is biased towards frequencies of higher amplitude, since they contribute higher to the feedback signal. Averaged over many SLM segments, these are the frequencies in the center of the Gaussian spectrum of the laser, resulting in a narrowing of the resulting spectrum. We investigated the dependence of this effect on the number of segments N by a numerical simulation, which generates random spectra based on the parameters of our experiment. These spectra were Fourier transformed and their sum optimized in time to mimic our experimental optimization procedure. For a low number of segments, the spectral amplitude and phase is dominated by the randomness from the scattering. For an increasing number of segments, the optimized pulses exhibit an increasingly smooth Gaussian amplitude and a flat spectral phase, with a bandwidth narrower than the input spectrum. The resulting pulse duration converges to 115 fs, in perfect agreement with our experiments. The method is capable of creating bandwidth-limited pulses, but since it is based on linear interferometry, the optimization can be equally applied to adapt other pulse shapes or likewise to compensate material dispersion present in the optical path.

The time-integrated intensity (energy) of the pulse with the highest peak depicted in Fig. 2 is 13.5 times higher than the energy of the average nonoptimized pulse. In addition to the temporal optimization, overall more light is transmitted into the detected channel, demonstrating that the scattered light is controlled spatially and temporally. A SLM alone, without a random scattering medium, offers only spatial control, while frequency domain pulse shaping techniques [29] provide temporal control only. Our method exploits the mixing of spatial and temporal degrees of freedom by the random medium [13], to control the transmitted light in two spatial and one temporal dimension by only controlling spatial degrees of freedom on the two-dimensional SLM. On the one hand, the conversion of

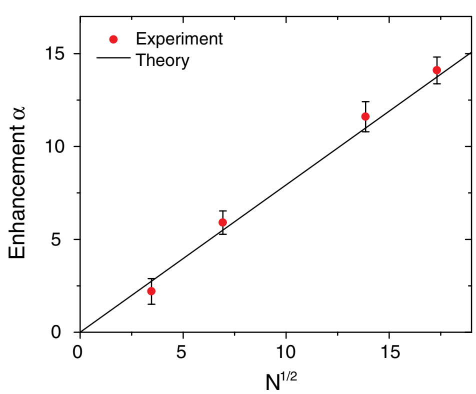

FIG. 4 (color online). Average amplitude enhancement α (dots) as a function of the square root of the number of segments N. The solid line is given by the expected $\alpha = 0.90(\frac{\pi}{4}N)^{1/2}$, without any free parameter.

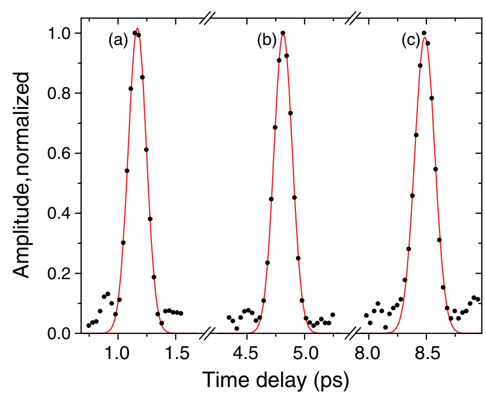

FIG. 5 (color online). Detailed cross-correlation scans after optimization with $N = 300$ segments around the time delays $\tau_{\mathrm{opt}} = 1.1$ ps (a), 4.8 ps (b), and 8.5 ps (c), together with a Gaussian fit (solid lines). The width (FWHM) of the peaks shows no significant dependence on the time delay, with an average of $\Delta\tau_{\mathrm{opt}} = (190 \pm 7)$ fs.

spatial degrees of freedom into temporal ones comes at the price of a speckle background, which on the other hand is easily outweighed by the enormous number of degrees of freedom provided by state-of-the-art SLMs. The large number of controllable spatial degrees of freedom is a great advantage over frequency domain pulse shaping techniques. Spatiotemporal control of the light field allows a far more generalized application of present coherent control schemes and marks a further step towards optical time reversal.

In the experimental realization presented here, we optimized the pulse front using linear interferometry as a feedback signal. The optimization of a nonlinear response, like second-harmonic generation, will also lead to a comparably optimized pulse [30]. Using second-harmonic emission from nanoparticles [14] or two-photon fluorescence from dyes such as fluorescent proteins [2] would enable focusing ultrashort pulses inside complex media such as biological tissue, in which the propagation of near-infrared light is dominated by multiple scattering [19]. Given the high signal-to-background ratio and the enhancement of energy delivered to the selected speckle spot, we envision that our method can improve approaches for selective cell destruction in tissue [31]. In view of its potential for sharp focussing, it has potential for nanofabrication, nanosurgery, and other micromanipulation techniques.

Up to now we have not discussed the spatial extent of the optimized pulse. We use a pinhole to select a single speckle spot in the Fourier plane of the sample for optimization. We know that transmitted fields in adjacent speckle spots are uncorrelated [32], from which we can conclude that the optimization is indeed limited to the selected area. An important future direction would be to investigate the spatial extent of the optimized pulse as a function of delay time. A combination with spatial scanning allows the measurement of the transmission matrix of the medium [33] in one temporal and two spatial dimensions. For Anderson-localizing samples [34], the size of the optimized speckle should be strongly time dependent [35].

In conclusion, we have experimentally demonstrated that spatial wave front shaping of a pulse front incident on a strongly scattering sample gives spatial and temporal control over the scattered light. Our approach provides a new tool for fundamental studies of light propagation and has potential for applications in sensing, nano- and biophotonics.

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