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Imaging in diffuse media with ultrafast degenerate optical parametric amplification

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We demonstrate the application of a subpicosecond optical parametric amplifier working at degeneracy to imaging in diffuse media. This optical parametric amplifier exhibits small-signal gains greater than 10^4 , thereby acting as a high-gain ultrafast amplifying gate. We have used it to construct the image of a grid pattern hidden behind 20 mean free paths of a highly diffusing solution of latex microspheres with a spatial resolution of 200 μm .

Transillumination imaging has generated much interest in recent years because of its potential as a diagnostic tool in medicine. It is hoped that alternative techniques to x-ray imaging that are nonionizing will be developed for applications in such areas as the early detection of breast cancer. Because human tissue is a highly scattering medium, there is a high probability that a photon being transmitted through a significant thickness of tissue will be deviated from its original path. However, photons that are either not deviated (called ballistic or unscattered photons) or little deviated (quasi-ballistic photons) from their original trajectory are potentially useful for detecting variations in the internal structure of the object. Therefore it is necessary to detect selectively the ballistic and quasi-ballistic photons with respect to the diffuse photons. Time-of-flight methods with high temporal resolution allow one to distinguish between the early-arriving ballistic and quasi-ballistic photons and the late-arriving diffuse photons.¹

Various methods of time-resolved imaging have been proposed as a means of transillumination imaging in diffuse media. These techniques have involved the use of a streak camera² or a picosecond Kerr gate¹ or have been based on such methods as heterodyne detection,³ second-harmonic cross correlation,⁴ and Raman amplification.⁵ In this Letter we show how an optical parametric amplifier (OPA) used as an ultrafast amplifying gate can be used to amplify primarily ballistic photons selectively and to construct a one-dimensional image of a grid pattern hidden behind 20 mean free paths of a diffuse solution of latex microspheres.⁶

The application of parametric amplification to the subpicosecond time resolution of weak signals has been demonstrated by Hulin *et al.*⁷ Parametric amplification, also referred to as difference-frequency generation, is particularly interesting not only because it is a process that can exhibit very high gains but also because the time during which amplification occurs is limited by the duration of the pump pulse. For subpicosecond pump pulses and small signals, an OPA can act as an ultrafast amplifying gate with gains exceeding 10^4 .

The setup for our OPA is shown in Fig. 1. The pump pulses were provided by second-harmonic generation of the pulses emitted from a Ti:sapphire laser chain. The laser chain, which was constructed in our laboratory, is similar to the one described by Squier *et al.*⁸ The Kerr-lens mode-locked Ti:sapphire oscillator, pumped by a cw argon laser, produced 2-nJ sub-100-fs pulses at 780 nm and at a repetition rate of 85 MHz. These pulses then were amplified by a factor of 10^6 through the now-standard technique of chirped-pulse amplification.⁹ First they were stretched to 200 ps in a single-diffraction grating stretcher working at Littrow incidence. The pulses then were amplified at 20 Hz in a Ti:sapphire regenerative amplifier with output pulse energies typically near 6 mJ. Finally they were recompressed in a two-grating Littrow incidence compressor. Losses in this compressor reduced the output pulse energy to near 2 mJ. A 3-mm β -barium borate (BBO) crystal (type I) acted as the frequency doubler to generate the 300- μJ pump pulses, and the undoubled portion of the 780-nm pulses was separated from these pump pulses to be used as the signal to probe the diffusive medium (see Fig. 1). The signal transmitted through the diffusive medium was temporally and spatially recombined with the pump in a second 3-mm BBO crystal (type I) oriented for collinear amplification at degeneracy. A delay line allowed one to adjust the relative arrival times of the pump and signal pulses in the OPA. In this way, it was possible to amplify the various portions of the transmitted signal pulses selectively while precompensating for the large group-velocity mismatch experienced by the signal and pump pulses in the second crystal. The amplified signal was detected by a silicon photodiode.

To test the limits of the system, we initially used neutral-density filters in place of the diffusive medium. We found that the degenerate OPA allowed us to detect, with a signal-to-noise ratio of 1.3, a signal that had passed through an optical density of 9. This corresponded to a 1-pJ seed pulse and an amplification of greater than 10^4 .

The gain of the OPA was limited by the combined effects of group-velocity dispersion and group-velocity

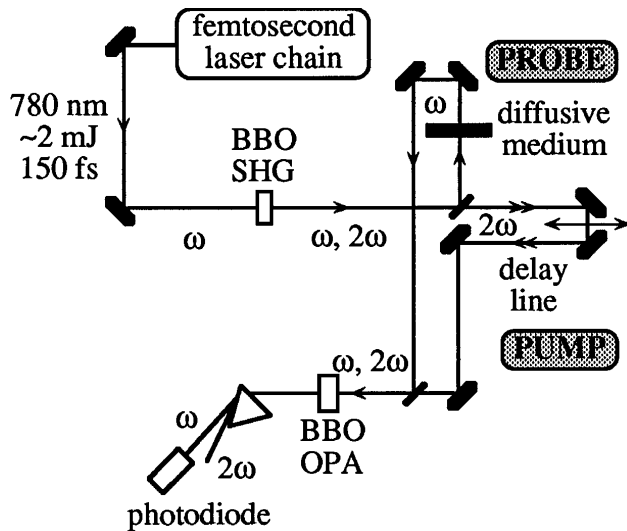


Fig. 1. Setup for the ultrafast degenerate OPA. The first crystal (BBO SHG) is used for the second-harmonic generation of the 780-nm pulses. The second crystal (BBO OPA) is the parametric amplifier oriented for degeneracy. ω refers to the 780-nm probe pulses and 2ω to the 390-nm pump pulses.

mismatch in the nonlinear crystals. To optimize the gain, we adjusted the grating separation in the compressor to precompensate for the group-velocity dispersion. It must be understood that the choice of a signal pulse at 780 nm with a pump pulse at 390 nm is unfortunate in this respect, because the group-velocity mismatch between the pump and signal is 200 fs/mm of BBO crystal. However, it should be noted that this signal wavelength is ideal for imaging biological tissues, because it falls in the so-called therapeutic window of low absorption in biological tissues.

To simulate the diffusing properties of tissue, we used a solution of latex microspheres (provided by Dow France) as our diffusive medium. The scattering cross section of this solution, $1.3 \times 10^{-11} \text{ cm}^2$, could be determined from Mie theory on the basis of the microsphere diameter (200 nm), the refractive index of latex (1.59), the refractive index of water (1.33), and the signal wavelength (780 nm). The number density of latex particles in the solution was $1.6 \times 10^{12} \text{ cm}^{-3}$, and the characteristic scattering length of the solution was approximately 0.05 cm. The scattering coefficient, μ_s , therefore was equal to 2 mm^{-1} . We note that this solution is less diffusive than real tissue, which has a scattering coefficient in the near infrared of 10 to 100 mm^{-1} .

Using our degenerate OPA, we detected a signal that traversed 1 cm (20 mean free paths) of the latex solution and measured the OPA signal as a function of the delay introduced to the pump pulses. The incident energy of the signal pulses on the latex solution was $100 \mu\text{J}$, and the pulses were focused to approximately $80 \mu\text{m}$. The pseudocorrelation of the pump and signal pulses (see Fig. 2) had a pulse width of 450 fs, indicating that the photons detected were primarily ballistic. The selectivity against the diffuse photons can be understood by considering the high angular selectivity of the OPA: these photons are

unlikely to enter the OPA crystal at exactly the right angle for collinear phase matching. It is, however, unfortunate that the angular selectivity seems to be so great that not even the quasi-ballistic photons can be amplified.

The proportion of unscattered (or ballistic) photons transmitted through 20 mean free paths of a diffuse medium is $e^{-20} (= 2 \times 10^{-9})$. Because the incident signal on the diffusing medium was $100 \mu\text{J}$, the energy of the transmitted signal was only 0.2 pJ. If we assume that the amplification of the OPA was of the order of 10^4 , the signal to be detected was approximately 2 nJ.

The fundamental limitation of our system is the parametric fluorescence that is generated on axis. The highly diffuse photons do not contribute to the noise level and are lost in the geometrical arrangement of the setup. Although the detection of purely ballistic photons is ideal for maximum spatial resolution, it is unrealistic for thick tissue samples, because the probability of a photon's being transmitted without deviation through a significant thickness of tissue is almost negligible. In fact, real tissues have not only a very high scattering coefficient, of $10\text{--}100 \text{ mm}^{-1}$ in the near infrared, but also a high anisotropy (or mean cosine of scatter), g , where g is 0.8–0.95. The transport coefficient (assuming negligibly small absorption) therefore can be as small as 0.5 mm^{-1} . Thus one can calculate that the proportion of unscattered light traversing 2 mm of soft tissues is less than 2×10^{-9} , while the proportion of scattered light traversing the same thickness potentially could be as high as 0.37! It is therefore important to consider the detection of the more numerous quasi-ballistic photons with some loss in spatial resolution. Currently we are working to reduce the angular selectivity of our apparatus to be able to detect these quasi-ballistic photons. By doing so, we should be able to extend the imaging capabilities of the OPA to beyond 20 mean free paths.

We then constructed point by point a one-dimensional image of a grid hidden behind 20 mean free paths of the latex solution. The solution was placed in a glass container with interior

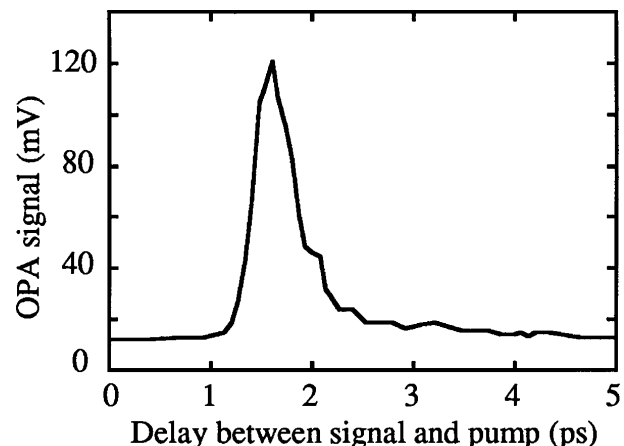


Fig. 2. OPA signal as a function of the delay between the signal and pump pulses. The FWHM is 450 fs, suggesting that primarily ballistic photons are detected.

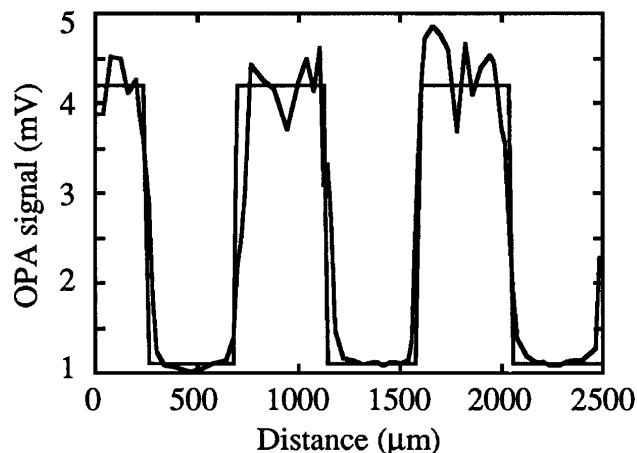


Fig. 3. Image of a grid hidden behind 20 mean free paths of latex solution. The signal-to-noise ratio is 4, and the resolution is $200 \mu\text{m}$.

dimensions of 10 mm (width) by 10 mm (thickness) by 40 mm (height), and the grid was placed on the exit face of the sample. The image that was constructed is shown in Fig. 3. One can see that the grid was detected with a signal-to-noise ratio of 4 and with a spatial resolution of $200 \mu\text{m}$. If the detection method is limited to the detection of purely ballistic photons, the spatial resolution should be determined solely by the spot size of the pulses in the diffuse medium (approximately $80 \mu\text{m}$) because, by definition, these photons follow a direct path. It should be noted that the placement of the grid behind the solution is not appropriate for the demonstration of imaging with scattered photons. In fact, high-resolution imaging can be achieved with such a positioning of the grid, regardless of the degree

of scatter. A more realistic test of the resolution limits of a detection method that uses scattered photons would require that the grid be placed within the scattering medium. However, because only unscattered photons were detected by our system, the exact placement of the grid was not critical.

In conclusion, we have demonstrated the application of a subpicosecond degenerate OPA for imaging in diffuse media. This OPA acted as an ultrafast amplifying gate with gains of greater than 10^4 for small signals. With it, we have constructed the image of a grid pattern with a spatial resolution of $200 \mu\text{m}$, behind 20 mean free paths of a solution of latex microspheres. We hope to improve the range of this imaging method by detecting the quasi-ballistic photons.

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