

Separation of coherent and incoherent nonlinearities in a heterodyne pump–probe experiment

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Abstract: We demonstrate that the transient coherent nonlinearity (coherent artifact) affecting the pump-probe response of semiconductor optical amplifiers can be experimentally separated from the incoherent transient. The technique is based on measuring the mirror component of the coherent artifact which is a background-free four-wave mixing signal at a different frequency with respect to the transmitted probe in a heterodyne detection scheme. Measurements on amplifiers of different length reveal strong deviations from the commonly expected symmetric shape of the coherent artifact in case of long waveguides.

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It has been recognized already for some time that in a pump–probe experiment, coherent interactions between the pump and probe pulses affect the probe transmission for small pump-probe relative delay times. For the geometry where pump and probe cross

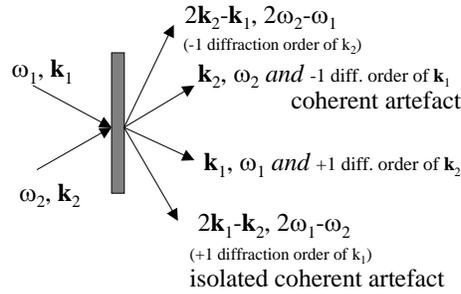


Fig. 1. Scheme of a diffraction geometry experiment in the spatial selection geometry. Along the probe direction \mathbf{k}_2 the first diffracted order of the pump is also present (coherent artifact). The diffracted mirror component is along the background-free direction $2\mathbf{k}_1 - \mathbf{k}_2$.

at an angle, the so-called coherent artifact is described as the pump radiation which is scattered into the probe beam by the spatial grating induced in the medium from the interference of the coherent pump-probe pulses [1, 2]. In the case of co-polarized transform-limited pump-probe beams, the effect of the coherent artifact on the probe transmission is estimated to be a symmetric function of the delay between the two pulses and equal in magnitude to the incoherent contribution at zero delay. Moreover, it is proportional to the pulse intensity autocorrelation if the third-order susceptibility is a step function in time [1, 2]. For non-transform limited pulses, the presence in the coherent artifact of an asymmetric contribution related to the real part of the susceptibility, i.e. to a refractive index grating, was also discussed [2]. Recently, a new pump-probe technique where both beams are co-polarized and co-parallel was demonstrated using a heterodyne detection scheme, suitable for experiments in active waveguides such as semiconductor optical amplifiers (SOAs) [3]. A theoretical treatment of the coherent artifact in this geometry was given in [4]. There, the role of the gain dispersion of the active medium (spectral artifact) on the coherent artifact was also evaluated. If the spectral gain slope is small, or the waveguide is very short, the general results mentioned above are recovered. A finite gain slope is shown to alter the shape of the coherent artifact from a symmetric signal with respect to the delay time into an asymmetric shape for the separate gain and refractive index changes [4].

Experimentally, the coherent and incoherent part in the change of the probe transmission are overlapped, which makes it difficult to extract information on the intrinsic dynamics of the investigated material at early times from a co-polarized pump-probe experiment. In order to separate the coherent from the incoherent contributions, comparison between co-polarized and cross-polarized experiment is often used, under the assumption that the interference effects vanish for cross-polarized beams [1]. Alternatively, the simple procedure that the coherent artifact is half of the incoherent signal and proportional to the intensity autocorrelation was used to correct the results from a pump-probe experiment [5]. In this article we propose a method to directly measure the coherent signal *separately* from the incoherent part. We present results that quantify the magnitude of the coherent artifact in two amplifiers of different length.

Let us first recall the different diffracted nonlinear signals in a grating-induced experiment in the usual transmission geometry where pump and probe cross at an angle (spatial-selection geometry) [6]. This is depicted in Fig. 1. The pump (probe) beam has the direction given by the wavevector \mathbf{k}_1 (\mathbf{k}_2), and the optical frequency ω_1 (ω_2). Along the zero order direction (i.e. the direct transmission) of the probe beam the first diffracted order of pump beam in the pump-probe induced grating is also present, degenerate with the probe frequency. This is the coherent artifact. The mirror component

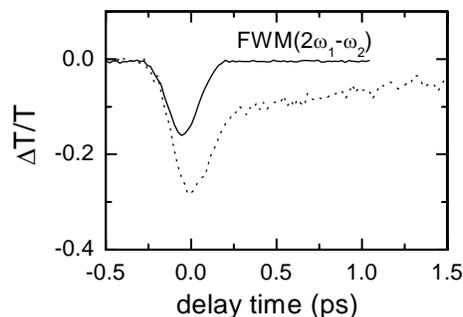


Fig. 2. Measured differential transmission (dotted) and coherent artifact (solid) in a $100\mu\text{m}$ -long SOA in the gain region.

of this diffraction, with respect to the zero-order transmitted pump, is along the direction $2\mathbf{k}_1 - \mathbf{k}_2$ and has the frequency $2\omega_1 - \omega_2$. Therefore, the coherent artifact can be measured separately from the transmitted probe, by detecting its mirror component which is at a different direction and frequency. The third-order coherent interaction giving rise to these diffracted signals is also called four-wave mixing (FWM)[6].

In a heterodyne detection scheme, pump and probe are co-propagating along the waveguide, and are distinguished by a small shift of the optical frequencies in a quasi-degenerate scheme[3]. Typically, acousto-optic modulators are used to provide a frequency shift in the radio frequency domain. The analogy of this scheme with the spatial selection geometry has been extensively discussed[4]. In this case the interference of pump and probe gives rise to a modulation of the gain and refractive index of the material in time rather than in space. This generates frequency sidebands to the pump and probe signals, in analogy with the different diffraction directions in Fig. 1. The sideband to the pump frequency which is degenerate with the probe signal is the coherent artifact and its mirror sideband can be detected background-free at a different frequency[4]. It was already pointed out experimentally[7, 8] how the heterodyne technique can be used to measure background-free four-wave mixing signals in SOAs. However, the coherent artifact was not addressed in these experiments.

In this work, we have used a heterodyne detection scheme to perform a pump-probe experiment on an InGaAs/InGaAsP multiple-quantum-well semiconductor optical amplifier operating at $1.5\mu\text{m}$ at room temperature. Pump and probe were co-polarized in the TE mode of the waveguide. The laser source was the idler of an optical parametric amplifier providing ~ 150 fs pulses at 300 kHz repetition rate. We have recently demonstrated the feasibility of a heterodyne setup with a low-repetition laser source of high amplitude noise[8]. The pump and probe pulses were nearly transform-limited, by compensating their linear chirp with the use of a pulse shaper[9]. Using cross-correlated frequency resolved optical gating we inferred Gaussian pulse shapes with a bandwidth product of 0.44. In our detection scheme the signal is measured by a lock-in referenced by an electrical mixing of the laser repetition rate and the acousto-optic modulator frequencies used in the setup[8]. We can easily switch between detecting the transmitted probe or the FWM signal at $2\omega_1 - \omega_2$ by adjusting only the lock-in reference while *keeping the experimental conditions unchanged*.

In Fig. 2 the differential transmission is shown (dotted line) as a function of the pump-probe delay time on a $100\mu\text{m}$ -long device. The pump pulse leads the probe at positive delays. At the used bias current of 35 mA a small signal gain of 6 dB was measured. The center optical frequency of the laser pulses was at $1.52\mu\text{m}$, close to the maximum of the amplified spontaneous emission spectrum. The pump energy per pulse at the input of the device was 0.26 pJ. The dynamics of the differential transmission shows an

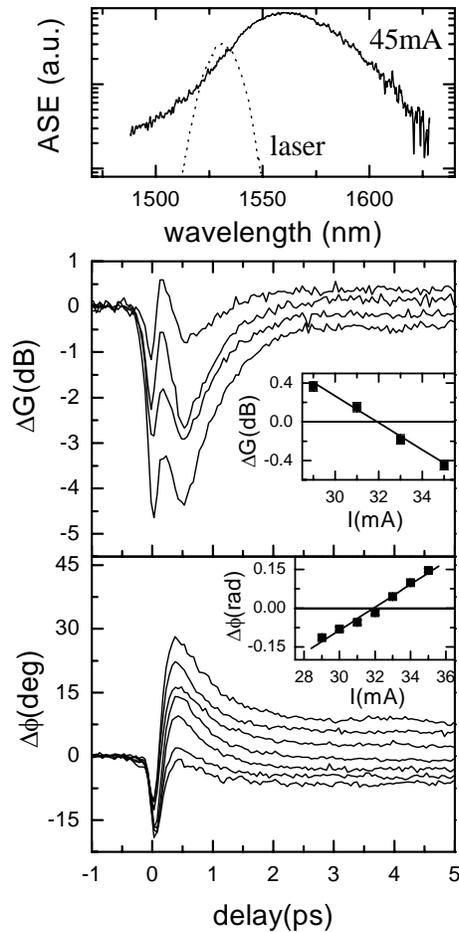


Fig. 3. Upper: amplified spontaneous emission in a 1mm-long SOA and pulse laser spectrum used. Lower: Gain changes (in dB) and probe phase changes versus the pump-probe delay, at different bias currents around transparency. In the inset, the values of the long-lived leftovers versus the bias current are shown. The values change sign, going from absorption to gain of the device, and cross zero at transparency.

initial negative transient with an ultrafast recovery comparable to the pulse duration, and a slower recovery over several hundred of femtoseconds. The ultrafast transient is usually attributed to spectral hole-burning, and the slower transient to carrier heating dynamics in SOAs[5]. In general, the occurrence of the coherent artifact, that overlaps the initial transient, makes it difficult to quantitatively estimate the spectral hole-burning component. For a grating formed by a modulation of the absorption coefficient, i.e., related to the imaginary part of the susceptibility, the scattered pump radiation is in phase with the transmitted probe. Thus, the amplitude of the FWM signal at $2\omega_1 - \omega_2$ fully quantifies the coherent artifact, if the scattered pump from the refractive index grating is negligible[2]. The solid line in Fig. 2 is the measured FWM signal at $2\omega_1 - \omega_2$ divided by the probe transmission signal without the pump, plotted as a negative transient. This is the *measured* contribution to the differential transmission given by the coherent artifact. This contribution is half of the transmission change at zero delay, and has a delay dependence like the pulse autocorrelation with a slight shift to negative delay, similar to expectations[4].

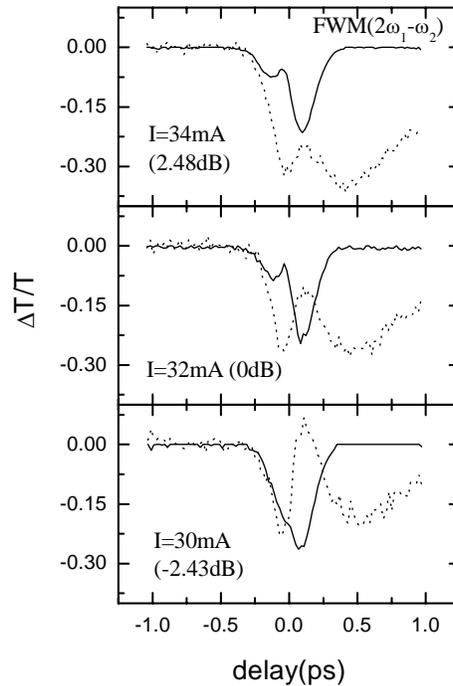


Fig. 4. Differential transmission (dotted) and coherent artifact (solid) in a 1mm-long SOA at different bias currents as indicated. The small signal gain (excluding internal waveguide losses) is also indicated.

In order to further explore the role of the coherent artifact in a pump-probe experiment, and its interplay with spectral artifacts, similar measurements were performed on a multiple quantum well SOA of 1 mm length, around the transparency current. In this case the spectrum of laser pulses probes a high gain slope, as shown in the top part of Fig. 3 where the laser spectrum is shown together with the amplified spontaneous emission from the device. The measured differential transmission around transparency current is shown in the lower part of Fig. 3. Due to the high propagation losses from the long waveguide, a high input pump energy (~ 4 pJ) was necessary to achieve an output signal with good signal-to-noise ratio. Both amplitude and phase changes of the transmitted probe are shown, corresponding to gain and refractive index changes[5]. In particular, the gain in dB deduced from the transmission change is indicated[8]. The long-lived leftovers of both gain and phase changes show the transition from absorption to gain of the device by changing their sign[5] (see insets of Fig. 3), and allow to infer a transparency current of 32 mA and an alpha parameter of 2.8. Similar to the case of the $100\mu\text{m}$ -long device, the dynamics of the differential transmission shows a recovery over several hundred of femtosecond due to carrier heating. However, at short positive delay times the gain dynamics are more complex than what measured in the short device, with the occurrence of strong oscillations. It was shown that spectral artefacts have an important role in explaining these oscillations[4]. However, since also the coherent artifact overlaps to the initial transient, it is interesting to experimentally measure its role in this case.

In Fig. 4 the differential transmission (dotted line) and the isolated coherent contribution (solid line) are shown for the 1 mm long SOA at three different bias currents, corresponding to gain, transparency and absorption (\sim of the device as indicated). The delay dependence of the coherent transient never follows the pulse autocorrelation but

always shows a double structure. Moreover, the coherent contribution is close to half of the transmission change at zero delay only in the gain case. These results show that the coherent transient nonlinearity, which overlaps the incoherent differential transmission, can have a complicated delay dependence, and might not correspond to half of the transmission change at zero delay. In the observed non-trivial delay dependence of the coherent transient, both higher order spectral artifacts and/or nonadiabatic dynamics of the material polarization[4] can play a role. Note also that the occurrence of a non-zero coherent transient at transparency indicates that coherent nonlinearities additional to those induced by a density grating are present, such as two-photon processes. In fact, two photon absorption is also observed in the measured phase dynamics around transparency, resulting in an initial negative transient[5]. The detailed understanding of this complicated scenario is, however, beyond the scope of this article.

In conclusion, we have demonstrated that the coherent transient affecting the nonlinear incoherent response in a pump-probe experiment on semiconductor optical amplifiers, known as coherent artifact, can be measured separately, and thus its isolate contribution can be experimentally estimated. This is achieved by measuring the mirror component of the coherent artifact which is a background-free four-wave mixing signal at a different frequency with respect to the transmitted probe in a heterodyne detection scheme. We find that the dependence of the coherent artifact on the pump-probe relative delay agrees with expectations in a short waveguide while deviates significantly in a long amplifier, with a shape well beyond the pulse autocorrelation profile. These findings have important implications for the analysis and understanding of the ultrafast dynamics in semiconductor active waveguides.