Adaptive pulse compression by two-photon absorption in semiconductors

U. Siegner

Physikalisch—Technische Bundesanstalt, Bundesallee 100, D-38116 Braunschweig, Germany

M. Haiml, J. Kunde,* and U. Keller

Swiss Federal Institute of Technology Zurich, Institute of Quantum Electronics, ETH Honggerberg-HPT, CH-8093 Zurich, Switzerland

Received September 6, 2001

We investigate the adaptive optimization of broadband laser pulses, using a closed-loop learning algorithm in which the merit function is derived from two-photon absorption in semiconductors. Photoluminescence experiments with CdS thin films and photocurrent measurements of a GaAsP photodiode have been performed. The experimental data demonstrate that reliable and accurate pulse compression to the bandwidth limit can be achieved, unperturbed by nontrivial phase effects. Therefore two-photon absorption proves to be an easy-to-implement alternative to second-harmonic generation for the compression of broadband laser pulses. © 2002 Optical Society of America

OCIS codes: 320.5520, 320.7130.

Adaptive feedback learning algorithms were originally proposed as a tool to control the interaction between light and matter.[1] Soon, however, it was realized that adaptive feedback control could also be used to improve the generation of ultrashort laser pulses.[2] Since then, several groups of researchers have experimentally demonstrated adaptive compression of ultrashort pulses from mode-locked lasers[3–5] and chirped-pulse amplifiers.[6–8] Recently, adaptive feedback control was utilized to generate 7-fs pulses with an optical parametric amplifier[9] and to optimize pulse propagation in optical fibers.[10] In adaptive feedback experiments, an optimization algorithm directs the shaping of optical pulses to maximize a merit function. The published experimental results were obtained by use of second-harmonic generation (SHG) signals as the merit function for pulse compression.[3–10] This approach has been validated by theoretical studies[2] and can be considered an established method. With broadband pulses, however, SHG has its difficulties. Proper phase matching and a constant conversion efficiency have to be ensured over a large spectral range to prevent spectral filtering of the broadband pulses.[11,12] Therefore, thin SHG crystals, which are costly and difficult to produce and yield only small SHG signals, have to be used. As a consequence, other ways to derive a merit function in adaptive pulse compression experiments are desirable.

Two-photon absorption (TPA) is a process that depends on the pulse intensity and, therefore, on the temporal width of the pulse for a fixed pulse energy. At first glance, one would expect the highest TPA signal for the shortest, bandwidth-limited pulse. When this holds true, the TPA signal can be used as a merit function for adaptive pulse compression.[13] However, nontrivial phase effects can occur in TPA experiments that will preclude the use of TPA signals as merit functions. In atomic systems the TPA probability for non-bandwidth-limited pulses with an asymmetric spectral phase is as high as that for bandwidth-limited pulses (assuming a fixed pulse energy and power spectrum).[14,15] Moreover, if a real intermediate state is involved in the TPA process, non-bandwidth-limited pulses with appropriate spectral phases can give rise to even stronger TPA than can bandwidth-limited pulses.[16]

So far, there has been no experimental investigation of which we were aware of whether nontrivial phase effects can occur in TPA experiments with semiconductors. The theories in Refs. 14–16 apply to noninteracting two-level or multilevel atoms and cannot be applied to strongly Coulomb correlated semiconductors[17,18] in a straightforward way. In particular, a semiconductor cannot be adequately described as an inhomogeneously broadened ensemble of independent two-level systems, for which nontrivial phase effects can be ruled out on theoretical grounds.[15]

In this Letter we show that the TPA signal in semiconductors is maximized by the shortest possible bandwidth-limited pulse and therefore can be used as a merit function for adaptive pulse compression. We performed photoluminescence experiments with CdS thin films and made photocurrent measurements of a GaAsP photodiode. The results show that reliable and accurate pulse compression to the bandwidth limit is achieved, unperturbed by nontrivial phase effects. Because TPA does not require phase matching, it is an easy-to-implement alternative to SHG for adaptive compression of broadband laser pulses. Using photodiodes, we obtained a compact setup for measurements of the merit function.

Our experiments were performed at room temperature with a home-built titanium:sapphire laser, generating pulses with a 50–60-nm broad spectrum (full width at half-maximum) near 800 nm at a 91.5-MHz repetition rate. The pulses propagate through a four-prism sequence so a bias group-delay dispersion (GDD) can be imposed. The pulses are then passed through a 4-f pulse shaper[19] with a liquid-crystal spatial light modulator with 128 independent pixels[20] (SLM-256-NIR, Cambridge Research & Instrumentation) at the Fourier plane. The setup is capable of amplitude and phase shaping; however, only the spectral phase is modulated in the present experiments. The pulse shaping is directed by an evolution strategy that controls the phase difference between adjacent pixels to maximize the merit function. Choosing discrete crossover as the recombination mechanism,[21] we achieved convergence after fewer than 50 iterations. A detailed description of the pulse shaper and the optimization algorithm can be found in Ref. 21. The optimization algorithm showed excellent performance in earlier adaptive control experiments with semiconductor nonlinearities.[21,22]

To validate TPA-based adaptive pulse compression, we compare the phase that maximizes the time-integrated SHG signal and the phase that maximizes the TPA signal. If the two phases are equal, one can conclude that reliable and accurate pulse compression to the bandwidth limit is achieved by use of a TPA signal as a merit function. Here we used the fact that SHG optimization results in a bandwidth-limited pulse with a flat temporal phase.[2,7] A first experiment was performed on a polycrystalline CdS (bandgap, 2.42 eV, corresponding to 512 nm) thin film of 1.8-μm thickness. The sample was excited near 800 nm with an average laser power $P_a = 21.5$ mW. In this experiment, as well as in all our other experiments, the laser beam was focused to a spot with a 18-μm diameter. The photoluminescence (PL) from the sample was collected over a 7-nm-wide band centered at 515 nm. We verified that this PL signal scales with ${P_a}^2$ for a fixed pulse shape, as expected for TPA. The PL signal was then iteratively maximized by the optimization algorithm to yield the optimum phase. Likewise, the time-integrated SHG signal from a 15-μm-thick ammonium dihydrogen phosphate crystal was maximized at the laser power $P_a = 21.5$ mW.[23]

Figure 1 shows the optimum spectral phases found for TPA and SHG optimization for three settings of the prism sequence, along with the pulse spectrum.[24] From top to bottom, the bias GDD at 800 nm increases by 207 fs^2 from curve to curve. In all cases, the TPA and the SHG phases agree very well in the spectral range in which the power is sufficiently high for optimization of the phase. Figure 1 demonstrates that reliable adaptive pulse compression is achieved by use of TPA-induced PL signals as merit functions. This result implies that phase distortions, which are introduced by dispersive optical elements, are accurately compensated for by the optimization algorithm if TPA signals are used as merit functions.

In a second experiment, we chose a TPA-induced photocurrent (PC) signal as the merit function. The PC was generated in a commercial GaAsP photodiode (Hamamatsu G1115, peak sensitivity at 640 nm) with an average laser power $P_a = 10$ mW. As a result of the TPA process, the PC signal scales with ${P_a}^2$ for a fixed pulse shape under our experimental conditions, as we experimentally verified. Again, a SHG optimization experiment was performed for comparison (at $P_a = 7$ mW). The optimum phases found for TPA and SHG optimization are plotted in Fig. 2 for two settings of the prism sequence. The bias GDD at 800 nm increases by 207 fs^2 from top to bottom. The spectrum of the pulses, also shown in Fig. 2, differs slightly from that used in the PL experiment. In the independent PC experiment we found again that the TPA and the SHG phases agree well in the spectral range in which the power is sufficiently high for phase optimization. Figure 2 confirms that reliable and accurate adaptive pulse compression can be obtained by use of TPA signals from semiconductors as merit functions.

Next we compared the experimental results with theoretical predictions. For this purpose we subtracted the reference spectral phase from the optimum phases for ±207-fs^2 bias GDD, using the TPA data of Fig. 1. The experimental difference phases obtained in this way reflect the excess spectral phase that we introduced by changing the settings of the four-prism sequence. The experimental difference phases were compared with the phase differences that could be calculated following the theoretical treatment in Ref. 25.

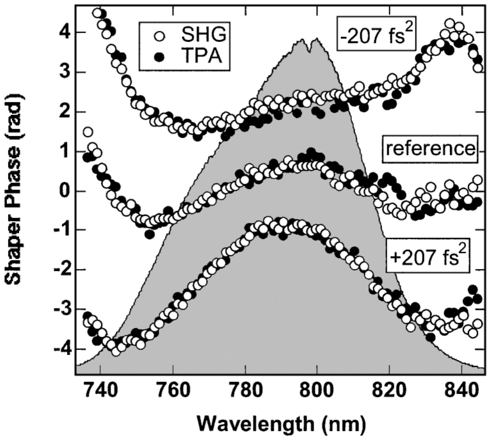

Fig. 1. Spectral phase that maximizes the TPA-induced photoluminescence signal from a CdS thin film and the SHG signal. Terms that are linear in frequency and constant terms have been subtracted. From top to bottom, the bias group-delay dispersion of the setup at 800 nm increases by 207 fs^2. Shaded area, power spectrum of the laser pulses. Number of iterations, 100.

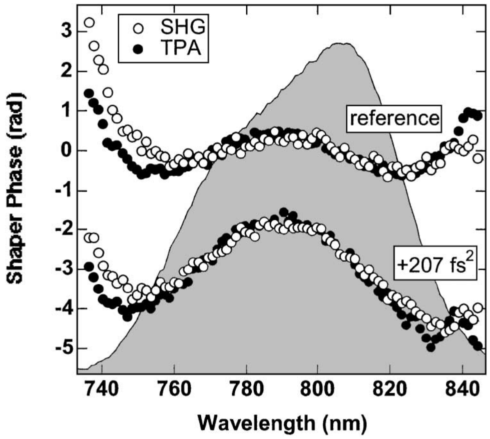

Fig. 2. Spectral phase that maximizes the TPA-induced photocurrent signal from a GaAsP photodiode and the SHG signal. Number of iterations, 50. All other conditions as in Fig. 1.

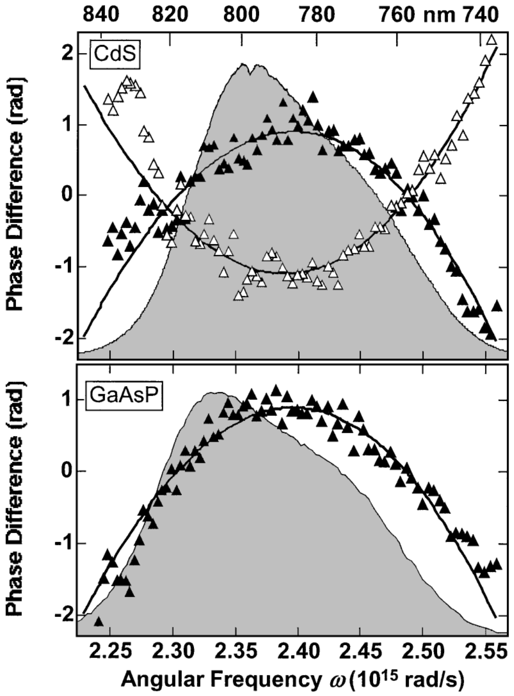

Fig. 3. TPA experiments: Optimum phase minus reference phase for the photoluminescence experiment with the CdS thin film and the photocurrent experiment with the GaAsP photodiode. Filled triangles, (phase for +207-fs^2 bias GDD) − (reference phase); open triangles, (phase for −207-fs^2 bias GDD) − (reference phase). The solid curves were calculated for a four-prism sequence; see text for details. Shaded areas, power spectra of the laser pulses.

More specifically, we calculated the spectral phases introduced by the various settings of the four-prism sequence that were used in the experiments. From these phases the phase differences between the prism settings were easily obtained. At the top of Fig. 3 the experimentally determined phase differences are compared with those predicted by theory for the experiment with the CdS thin film. Good agreement is found, confirming again that TPA-based adaptive pulse compression can accurately compensate for phase distortions introduced by dispersive optical elements. This conclusion was reconfirmed by analysis of the TPA experiment with the GaAsP photodiode. Following the procedure outlined above, we obtained the experimental and the theoretical phase differences shown at the bottom in Fig. 3. These data match well.

In summary, we have demonstrated that two-photon absorption in semiconductors can be utilized for the adaptive compression of ultrashort broadband laser pulses. Reliable compression to the bandwidth limit is achieved with this approach.

The authors thank A. Romeo and H. Zogg for growing the CdS thin film. U. Siegner's e-mail address is uwe.siegner@ptb.de.

*Present address, Centre Suisse d'Electronique et de Microtechnique, CH-6055 Alpnach, Switzerland.

References

1. R. S. Judson and H. Rabitz, Phys. Rev. Lett. **68,** 1500 (1992).
2. D. Meshulach, D. Yelin, and Y. Silberberg, Opt. Commun. **138,** 345 (1997).
3. D. Yelin, D. Meshulach, and Y. Silberberg, Opt. Lett. **22,** 1793 (1997).
4. T. Baumert, T. Brixner, V. Seyfried, M. Strehle, and G. Gerber, Appl. Phys. B **65,** 779 (1997).
5. E. Zeek, K. Maginnis, S. Backus, U. Russek, M. Murnane, G. Mourou, and H. Kapteyn, Opt. Lett. **24,** 493 (1999).
6. A. Efimov, M. D. Moores, N. M. Beach, J. L. Krause, and D. H. Reitze, Opt. Lett. **23,** 1915 (1998).
7. T. Brixner, M. Strehle, and G. Gerber, Appl. Phys. B **68,** 281 (1999).
8. E. Zeek, R. Bartels, M. M. Murnane, H. C. Kapteyn, S. Backus, and G. Vdovin, Opt. Lett. **25,** 587 (2000).
9. M. R. Armstrong, P. Plachta, E. A. Ponomarev, and R. J. D. Miller, Opt. Lett. **26,** 1152 (2001).
10. F. Omenetto, A. J. Taylor, M. D. Moores, and D. H. Reitze, Opt. Lett. **26,** 938 (2001).
11. A. M. Weiner, IEEE J. Quantum Electron. **QE-19,** 1276 (1983).
12. G. Taft, A. Rundquist, M. M. Murnane, I. P. Christov, H. C. Kapteyn, K. W. DeLong, D. N. Fittinghoff, M. A. Krumbügel, J. N. Sweetser, and R. Trebino, IEEE J. Sel. Top. Quantum Electron. **2,** 575 (1996).
13. In Ref. 7 it was mentioned that TPA in a SiC photodiode was used as a merit function. However, no experimental data were shown, and no careful analysis and validation of the method were reported.
14. D. Meshulach and Y. Silberberg, Nature (London) **396,** 239 (1998).
15. D. Meshulach and Y. Silberberg, Phys. Rev. A **60,** 1287 (1999).
16. N. Dudovich, B. Dayan, S. M. Gallagher Faeder, and Y. Siberberg, Phys. Rev. Lett. **86,** 47 (2001).
17. J. Shah, *Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanostructures* (Springer-Verlag, Berlin, 1999).
18. D. S. Chemla, in *Nonlinear Optics in Semiconductors I*, R. K. Willardson and E. R. Weber, eds., Vol. 58 of Semiconductors and Semimetals (Academic, Boston, Mass., 1999), p. 175.
19. A. M. Weiner, Prog. Quantum Electron. **19,** 161 (1995).
20. M. M. Wefers and K. A. Nelson, Opt. Lett. **20,** 1047 (1995).
21. J. Kunde, B. Baumann, S. Arlt, F. Morier-Genoud, U. Siegner, and U. Keller, J. Opt. Soc. Am. B **18,** 872 (2001).
22. J. Kunde, B. Baumann, S. Arlt, F. Morier-Genoud, U. Siegner, and U. Keller, Appl. Phys. Lett. **77,** 924 (2000).
23. In all our optimization experiments we divided the TPA or SHG signal by $P_a{}^2$ to account for long-time drift of the laser power.
24. Terms that are linear in frequency and constant terms were subtracted from the phases shown in this Letter, as they contain no relevant physical information in experiments with single pulses.
25. R. L. Fork, O. E. Martinez, and J. P. Gordon, Opt. Lett. **9,** 150 (1984).