Coherent two-photon absorption quenching of short laser pulses with phase–amplitude irregularities

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A phase and amplitude irregularity introduced into the spectrum of a short laser pulse by propagation through a weak narrow-line absorber results in strong suppression of continuum two-photon absorption at the peak frequency of the narrow-line absorber. © 2003 Optical Society of America

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1. INTRODUCTION

The propagation of intense short pulses in transparent media has been studied in great detail in materials such as silica glass[1] and in semiconductors such as GaAs.[2–10] The nonlinear interaction of a pulse with such a material is dominated by $\chi^{(3)}$ processes, and, when propagation is concerned, the dominant $\chi^{(3)}$ contributions correspond to two-photon absorption (TPA) and to nonlinear refraction (the Kerr effect). The Kerr effect is in fact related to two-photon transition amplitudes between continuum states (the valence and conduction band states)[3] as well as to Raman transitions and the optical Stark effect[4] through nonlinear Kramers–Krönig relations.[5] These and other $\chi^{(3)}$ processes affect the amplitude and the phase of the pulse as it propagates in the medium and strongly modify its temporal shape and spectral content.[1,11] In a different context, $\chi^{(3)}$ processes are sensitive to small changes in amplitude and phase of a pulse and are therefore implemented in characterization tools that permit reconstruction of the complex envelope functions of arbitrary laser pulses.[12,13]

Although the propagation of short pulses in glasses and semiconductors has been thoroughly investigated, and the crucial importance of having the exact pulse shape in this problem is recognized, relatively little effort has been dedicated to studying the consequences to propagation of pulse shaping. In particular, the ways in which shaping affects the nonlinear absorption in these materials have not been studied. This deficiency contrasts sharply with the considerable attention given to coherent control experiments, in which the possibility of manipulating the coherence properties of laser pulses is exploited as a means to control quantum processes such as chemical reactions,[14,15] one-photon transitions in two-level systems,[16] and phonon emission by photocarriers in semiconductors.[17,18] Coherent control of TPA was studied in two-level systems[19,20] by use of state-of-the art pulse-shaping techniques.[21] In this case, manipulation of the pulse modifies the quantum interference of all possible trajectories that lead to the same final state. The result is that the multiphoton transition probability is strongly altered and may even be completely suppressed.[19,20] It was shown, however, that inhomogeneous broadening of the optical transition tends to wash out this quantum interference effect.[20] This result may lead one to suspect that, because of the huge inhomogeneous broadening that characterizes the transitions between continuum states in semiconductors and glasses, similar phase-sensitive effects on multiphoton absorption in such materials would be strongly quenched. The extent of such quenching obviously depends on the time scale of the experiment, but, even in the limit of instantaneous response of the nonlinear material, the relative phases of the various spectral components of the pulse may determine the extent to which the components may contribute to the absorption.

In this paper we demonstrate how modifications of the phase and the amplitude of an incident pulse indeed strongly modify TPA of that pulse in a continuum nonlinear absorber, even in the limit of instantaneous nonlinear response. In the experiment we introduce phase and amplitude irregularities by letting the pulse propagate through a weak, narrow-line linear absorber. For semiconductors that exhibit strong TPA in the infrared the result is remarkable: Although the effect on the total nonlinear absorption in the material is minor, the spectrum of the transmitted pulse shows pronounced peaks centered about each of the irregularities in the incident spectrum. Surprisingly, the continuum nonlinear absorber becomes almost transparent at the peak frequencies of the narrow-line absorber, contrary to the expectation that TPA will have no preference for particular frequency components within the spectrum of the pulse. The main features of the experiment are reproduced by a model that takes into account the Kerr effect, TPA, and the complex spectral response function of the narrow-line linear absorber.

2. EXPERIMENT

We use high-energy ($\leqslant 2$-μJ), $\approx$50-fs pulses, generated by a Spectra-Physics OPA-800 optical parametric amplifier.

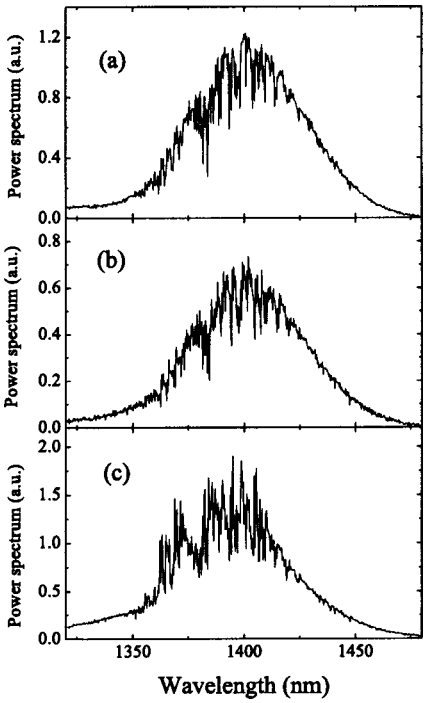

Fig. 1. Incident and transmitted spectra for InP, with the laser tuned to 1400 nm (300-g/mm grating): (a) incident spectrum, (b) transmitted spectrum at low input intensity (0.02-μJ pulse energy); (c) transmitted spectrum at high input intensity (2-μJ pulse energy).

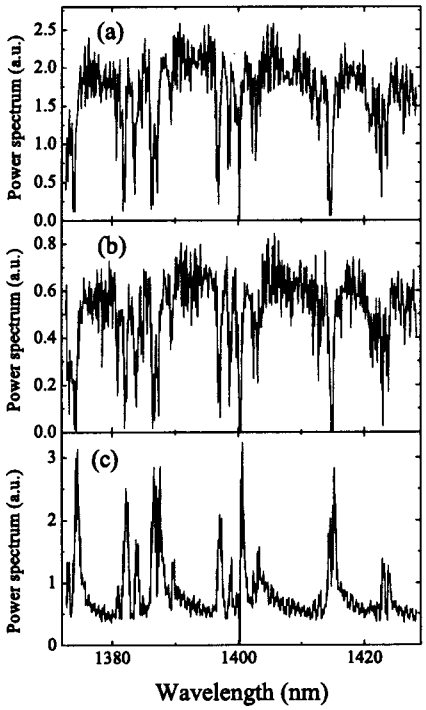

Fig. 2. Incident and transmitted spectra for InP, with the laser tuned to 1400 nm (1200-g/mm grating): (a) incident spectrum, (b) transmitted spectrum at low input intensity (0.02-μJ pulse energy), (c) transmitted spectrum at high input intensity (2-μJ pulse energy).

The pulses propagate $\approx$4 m before they are focused to an $\approx$250-μm spot on the sample. At infrared wavelengths that are characterized by narrow and relatively strong atmospheric absorption lines, a path length of $\approx$4 m is sufficient to introduce weak but measurable phase and amplitude irregularities to the pulse spectrum. The pulse then interacts with the nonlinear sample: a 500-μm-thick wafer of compensated GaAs or InP, polished on both sides, or a 6-mm-thick glass window. The spectrum of the transmitted beam is measured with a 50-cm spectrometer and an InGaAs detector array.

Experimental data obtained at 1400 nm with an InP sample are presented in Fig. 1. A strong band of atmospheric absorption is known to exist in this wavelength range,[22] and the effect of atmospheric absorption is clearly visible Fig. 1(a) in the form of sharp dips in the spectrum of the incident beam. The spectra of the transmitted beam for input pulse energies of 0.02 and 2 μJ are shown in Figs. 1(b) and 1(c), respectively (not to scale). Whereas at low intensities the spectrum of the transmitted beam is virtually identical to that of the incident beam, the transmitted spectrum at high intensities is markedly different: The series of sharp dips is transformed into an alternating series of peaks and dips. Importantly, the intensity dependence confirms that the changes seen in the spectrum are due to a nonlinear effect. The picture becomes clear when the same spectra are measured with higher spectral resolution (Fig. 2). Remarkably, every dip in the incident spectrum is transformed, at high pulse energies, into a peak in the transmitted spectrum. Similar data measured with a GaAs sample are presented in Fig. 3. The dotted and solid curves at the top of Fig. 3 are the spectrum of a high-intensity incident beam and that of the transmitted

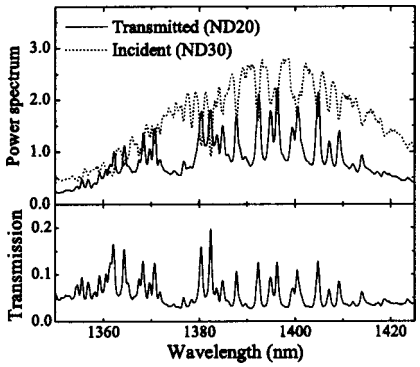

Fig. 3. Spectra for GaAs at high power (2-μJ pulse energy) with the laser tuned to 1400 nm (300-g/mm grating). Top, incident and transmitted spectra; bottom, normalized transmission. ND_x refers to the neutral-density filter used in each measurement.

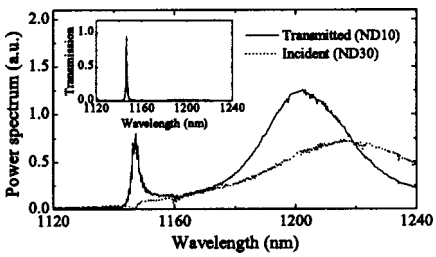

Fig. 4. Incident and transmitted spectra for GaAs at high power (2-μJ pulse energy), with the laser tuned to 1220 nm (300-g/mm grating). Inset, normalized transmission. ND_x refers to the neutral-density filter used in each measurement.

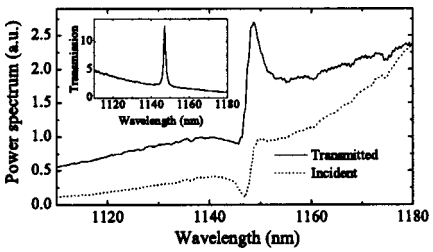

Fig. 5. Incident and transmitted spectra for glass at high power (2-μJ pulse energy) with the laser tuned to 1220 nm (300-g/mm grating). Inset, normalized transmission.

beam, respectively. The normalized transmission spectrum is shown at the bottom of the figure. The average transmission, $\approx$0.05, is consistent with the experimental parameters and the known TPA coefficient of GaAs at 1400 nm.[6] However, the transmission at each peak position is considerably higher, as great as 0.2.

The behavior described above is also observed in other spectral ranges where atmospheric absorption occurs. Figure 4 shows experimental data obtained with the laser tuned to 1220 nm with a GaAs sample. The dotted and solid curves are the spectrum of a high-intensity incident beam and that of the transmitted beam, respectively, and the normalized transmission spectrum is shown in the inset. In this case an isolated asymmetrical peak is clearly observed in the transmitted spectrum, at a wavelength of 1147 nm, at the position of a narrow dip in the incident spectrum. The normalized transmission at the peak is almost unity, much higher than the average transmission of $\approx$0.02. Remarkably, the continuum nonlinear absorber becomes almost transparent at the peak frequency of the narrow-line absorber.

The experimental data demonstrate that in the nonlinear regime the transmission of the semiconductor becomes frequency selective. This observation is contrary to the expectation that TPA will have no preference for particular frequency components within the spectrum of the pulse. In fact, all pair combinations of photon energies are expected to contribute equally in the case of a continuum absorber. However, it is the *interference* of all these two-photon transition amplitudes that determines the TPA coefficient at any frequency. This means that any explanation of the observed phenomenon must involve the phase of the laser field and not only its amplitude. Consequently, the complex response function of the medium through which the laser pulse propagates must be considered. This requirement concerns not only the spectrally dependent phase that the atmospheric absorbers impart to the laser field but also the complex nonlinear response. Therefore we must also consider the role of nonlinear refraction, i.e., the Kerr effect, which is the real counterpart of the (imaginary) TPA term in nonlinear susceptibility.[5]

To examine the relative roles of TPA and the Kerr nonlinearity in producing the experimental results we performed identical measurements with the glass sample. Here TPA is negligible, and the Kerr nonlinearity dominates. Figure 5 shows data for glass with the laser tuned to 1220 nm. The results are similar to those obtained with GaAs and InP, with the important difference that the normalized transmission is larger than 1 everywhere. The latter property is a trivial consequence of the spectral broadening of the pulse that is due to self-phase modulation (SPM).[1,23] From these data one might conclude that only the Kerr nonlinearity is important for the formation of peaks in the transmitted spectra: In analogy with SPM, the formation of a peak may then be interpreted as nonlinear frequency conversion near a phase and amplitude irregularity. However, the effects of SPM are strongly suppressed in the presence of TPA. Therefore the fact that the peak survives in the presence of TPA and actually dominates the transmission spectra of GaAs and InP is remarkable. As we shall see in what follows, *TPA alone can also form a peak in the transmitted spectrum*.

3. THEORY

To confirm that the Kerr nonlinearity and TPA are sufficient to explain the experimental data, we model our experiment in the following way: We define an input pulse with a Gaussian envelope function, $\exp(-4t^2/\tau^2)$, where τ is the pulse width. This envelope function is then multiplied, in Fourier space, by the complex transfer function of a Lorentzian absorption line, $H(\omega, \Delta) = \exp[-zP(\omega, \Delta)]$, where $P(\omega, \Delta)=C/[\Gamma - i(\Delta - \omega)]$.[24] Here ω is the frequency, Δ is the detuning, Γ is the linewidth, z is the propagation length through the absorber, and C is a constant. In the calculations presented here the parameters of the pulse and of the Lorentzian absorption line are fixed at $\tau = 50$, $\Delta = 0.05$, $\Gamma = 0.002$, $C = 0.002$, and $z = 1$. The complex spectral transfer function for the above parameters is depicted in Fig. 6. Figure 6(a) shows the amplitude, superimposed upon the input pulse spectrum, and Fig. 6(b) shows the phase. The effect of the linear absorber on the power spectrum of the pulse is simply the appearance of a narrow dip, as can be observed experimentally. The effect of the phase is best seen, however, in the time domain. In Fig. 7 we present the amplitude of the pulse after it propagates through the linear absorber [Fig. 7(a)], its phase [Fig. 7(b)], and the absolute square of its complex envelope function [Fig. 7(c) on a logarithmic scale]. We find that the propagation of the laser pulse through the linear absorber results in the formation of a weak exponential tail behind the pulse and that the phase varies rapidly within this tail. The tail has an intuitive explanation: It results from the polarization that the pulse induces in the linear absorber, which subsequently decays with a lifetime that is inversely proportional to the linewidth. Consequently the decay time of the tail trailing behind the pulse is the lifetime of the linear absorber, and the tail's frequency content corresponds to the absorber's frequency (the tail in fact represents the exponential impulse response of the linear absorber, which is simply the Fourier transform of its complex Lorentzian transfer function). The weakness of the tail makes it virtually undetectable in the experiment. However, as we discuss below, this tail is the clue to an intuitive understanding of the observed nonlinear behavior.

In calculating the effect of propagation through the nonlinear medium we assume that the nonlinear response of the medium is instantaneous. This assumption is valid for the experimental conditions, which involve

transitions between continuum states. It is a common assumption in the analysis of propagation phenomena[1] and considerably simplifies our model by permitting a straightforward integration of the nonlinear propagation equation in the time domain. The propagation equation that we use for the slowly varying amplitude of the pulse, $A(z, t)$, is derived from the one-dimensional nonlinear Schrödinger equation[1]: $\partial A/\partial z + (\alpha - i\gamma)|A|^2 A = 0$, where α and γ are the nonlinear propagation constants related to TPA and the Kerr effect, respectively. With $|A|$ normalized to unity at the peak of the pulse, α is calculated from the TPA coefficient α_2 (given in units of centimeters per gigawatt) through the relation $2\alpha l = \alpha_2 IL$,

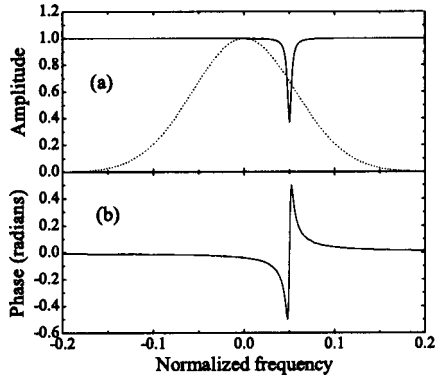

Fig. 6. Complex spectral transfer function of a Lorentzian absorption line. (a) Amplitude of the transfer function superimposed upon the input spectrum, (b) phase of the transfer function (the parameters are given in the text).

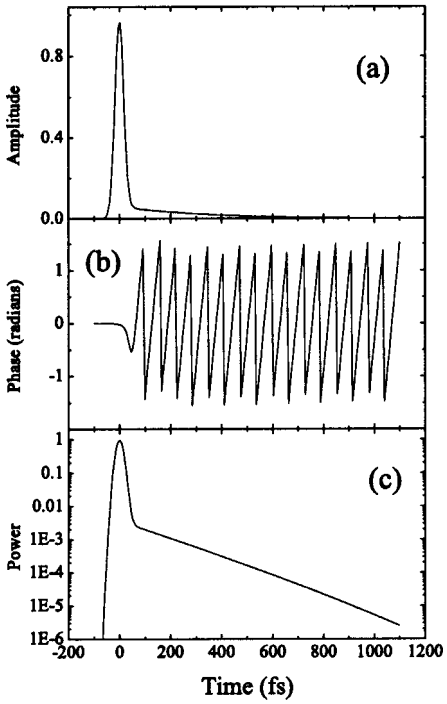

Fig. 7. Modified temporal shape of an input Gaussian pulse after it propagates through a linear absorber characterized by the transfer function shown in Fig. 6. (a) Amplitude of the modified pulse, (b) the pulse's time dependent phase. The absolute square of the complex envelope function is depicted in (c) on a logarithmic scale. Note the weak exponential tail trailing behind the pulse.

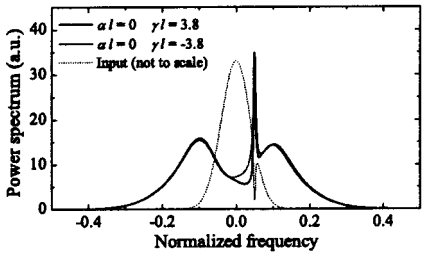

Fig. 8. Calculated transmitted power spectra for a pure Kerr medium ($\alpha = 0$). The dashed curve is the power spectrum at the input of the nonlinear medium.

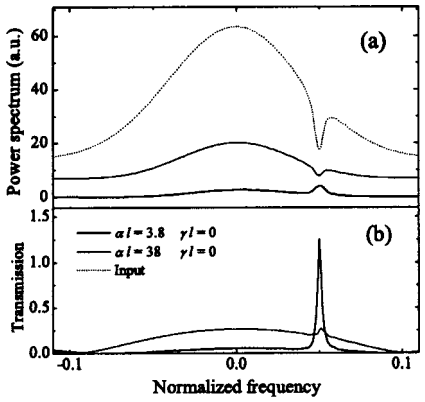

Fig. 9. Calculated spectra for a medium characterized by a purely dissipative nonlinearity ($\gamma = 0$). (a) Transmitted power spectra, (b) the corresponding normalized transmission curves. The dashed curve in (a) is the power spectrum at the input of the nonlinear medium. The curves in (a) are displaced vertically in steps of 7 a.u. to improve clarity.

where l is the number of iterations (i.e., propagation steps) in the calculation, L is the total thickness of the sample, and I is the power density, in units of watts per square centimeter. Similarly, the relationship between γ and the nonlinear index of refraction n_2 is $\gamma l = 2\pi n_2 IL/\lambda_0$, where λ_0 is the central wavelength of the laser. For simplicity, we neglect the variation of the nonlinear refraction and absorption coefficients over the pulse spectrum. (Although both parameters vary considerably in the wavelength range of our experiment,[2–10] the laser bandwidth is still small enough to allow us to make this first-order approximation, which considerably simplifies the calculations; as shown below, this simplistic approach is sufficient to describe the main features of the experiment.) Finally, using the dispersion coefficients from the literature, 1000 fs^2/mm for GaAs (Ref. 25) and 10.2 fs^2/mm for glass at 1147 nm,[1] we obtain dispersion lengths of 2.5 and 250 mm, respectively, for a 50-fs pulse. The propagation length through the nonlinear material in our experiment is therefore considerably shorter than the dispersion length for all the samples that we have used. This, and the fact that the experiment is performed in the limit of high laser fluences and short propagation lengths, justifies the neglect of dispersion in our model.

Next we present a parametric study of our model's predictions. In Fig. 8 we present the calculated output spectrum for a medium characterized by pure Kerr nonlinear-

ity without TPA. The spectral broadening that is due to SPM is clearly evident, and so is the appearance of a sharp peak in place of the dip in the input spectrum. Note that the peak appears for both signs of γ and that the sign determines the symmetry of the line shape: A change of sign only enhances one side of the resonance relative to the other. In Fig. 9 we show the calculated output spectrum for a medium characterized by a purely dissipative nonlinearity, where α is positive and $\gamma = 0$. Figure 9 demonstrates that TPA by itself can also form a peak in the transmitted spectrum. In this case, when the nonlinearity is weak or the propagation distance is short, the peak is seen only in the normalized transmission spectrum [Fig. 9(b)] but not in the transmitted power spectrum [Fig. 9(a)]. However, as αl increases, a peak also forms in the transmitted power spectrum and actually dominates the normalized transmission spectrum. In Fig. 10 we present data calculated for a nonlinear medium for which both TPA and the Kerr nonlinearity are significant. A comparison of the data in Fig. 10 with the data in Fig. 9 reveals that the Kerr nonlinearity strongly enhances the transmission at the resonance frequency. A comparison of the data in Fig. 10 and the data in Fig. 8 shows that TPA, as expected, suppresses the spectral broadening that is due to SPM. Also note that, to produce the same resonant transmission at the phase–amplitude irregularity, a larger γ is required in the presence of TPA (in spite of the fact that TPA alone can produce the resonant peak in the transmitted spectrum). As in Fig. 8, the sign of γ determines the symmetry of the line shape.

We now turn to a comparison of the model's predictions with the experiment. Figure 11 shows a calculated spectrum for parameters that correspond to Fig. 4 (GaAs). Good agreement with the experimental data is obtained for $\alpha l = 50$ and $\gamma l = -20$. These parameters are within

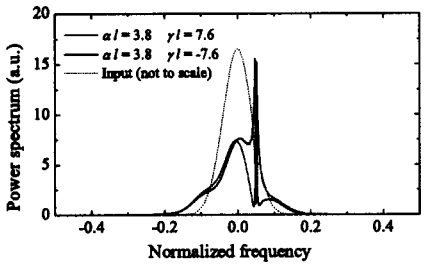

Fig. 10. Calculated transmitted power spectra for a medium with both nonlinear absorption and a nonlinear refractive index. The dashed curve is the power spectrum at the input of the nonlinear medium.

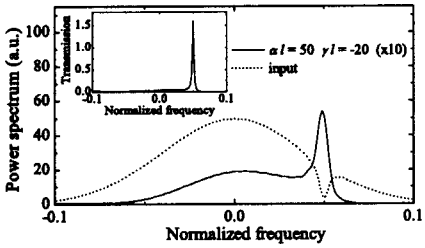

Fig. 11. Calculated spectra for the experimental parameters of Fig. 4. Inset, the normalized transmission.

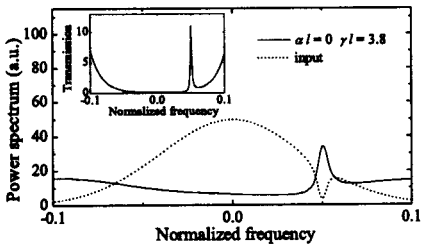

Fig. 12. Calculated spectra for the experimental parameters of Fig. 5. Inset, normalized transmission.

20% of the corresponding nonlinear constants of GaAs,[6,10] $\alpha_2 \approx 25$ cm/GW and $n_2 \approx -1.5 \times 10^{-13}$ cm^2/W at $\lambda_0 = 1.2\ \mu$m, assuming a power density of $\approx$70 GW/cm^2 (which corresponds to a pulse energy of 2 μJ, a pulse duration of 60 fs, and a beam waist of 250-μm diameter). In particular, the calculation reproduces the experimental average and peak transmissions, and the asymmetry of the calculated line shape is the same as in the experimental data for GaAs (note that Fig. 4 shows data as function of wavelength, whereas Fig. 11 shows data as function of frequency). For glass, $\alpha = 0$ and $n_2 \approx 3 \times 10^{-16}$ cm^2/W,[1] corresponding to $\gamma l = 6.3$. However, better agreement with the experimental peak transmission (Fig. 5) is obtained for a somewhat lower value of the Kerr nonlinearity, $\gamma l = 3.8$ (Fig. 12). Note that, in contrast to the data for GaAs, the asymmetry of the calculated line shape for glass does not reproduce the experimental data well. In fact, the asymmetry seems better for a calculation with $\gamma l = -3.8$. A possible explanation for these discrepancies is that some of our assumptions are not so good for glass as they are for GaAs and InP (in particular, the huge spectral broadening that is due to SPM that we observe in glass indicates that neglect of dispersion in this case is not completely justified). Nevertheless, the main feature of the experimental data, the high peak transmission, is reproduced well.

4. DISCUSSION

The calculations presented in Section 3 provide an insight into the origins of the behavior that we observe in the experiment. As discussed above, the propagation of a laser pulse through a linear absorber results in the formation of a weak exponential tail behind the pulse (see Fig. 7), which is the result of the polarization induced by the pulse in the linear absorber. This weak tail provides a clue to an intuitive explanation for the suppression of TPA at the frequency of a phase–amplitude irregularity. In the limit of instantaneous response of the nonlinear medium, the weak tail is not affected by TPA, whereas the main part of the pulse is strongly attenuated. Therefore the frequency content of the tail, which corresponds to the linear absorber's resonant frequency, goes through the nonlinear absorber without being attenuated by TPA. This intuitive interpretation explains the filtering effect that we observe in the spectral domain and is in agreement with the prediction of the model that the filtering effect will persist even when n_2 vanishes, i.e., when there is no self-modulation of the phase of the optical field in the

nonlinear absorber. This conclusion implies that the phase imprinted upon the pulse not only affects SPM but also coherently quenches TPA in a continuum absorber.

The intuitive interpretation above is appropriate only in special cases, however (e.g., for GaAs at a photon energy of ≈ 1 eV, where the sign of n_2 changes) and certainly cannot be invoked for glass, for which the Kerr effect is the dominant nonlinearity. The strong effect that we observe in glass actually shows that SPM by itself can also give rise to a frequency-selective enhancement of the transmission. In this case the effect can be understood as an interplay between the phase structure that was imprinted upon the pulse by the linear absorber and SPM in the nonlinear material. Although this interplay is not so transparent as the mechanism proposed above for a purely absorptive nonlinearity, we suggest the following intuitive picture: The exponential tail that forms behind the pulse varies slowly compared with the main part of the pulse, and therefore it is affected less by SPM. The frequency content of the tail is therefore locked and is not pushed to the wings of the spectrum. The actual transmission at the phase irregularity may then depend on details such as the detuning of the linear absorber from the center frequency of the laser, the signs and magnitudes of n_2 and the group-velocity dispersion, and the input intensity. Further experimental and theoretical investigations of this issue are necessary.

The explanation given above emphasizes the temporal shape of the pulse as a key to understanding the observed phenomena. Inasmuch as this temporal shape is clearly sensitive to the phase that is imprinted upon the spectrum, we conclude that the experimental results indeed demonstrate the sensitivity of the nonlinear interactions not only to the amplitude but also to the phase of the optical field. A complete understanding of the interaction of the field with the nonlinear material therefore requires that both the amplitude and the phase of the pulse be taken into account, and their interplay with the real and imaginary parts of the material's complex response function must be analyzed. Clearly, the phase modifications turn out to have important consequences, even when the nonlinear interaction is apparently sensitive only to the instantaneous intensity profile, which is only slightly modified by these phase modifications.

5. CONCLUSIONS

In conclusion, our experiment and theory show that small modifications to the phase and amplitude of a pulse may affect the nonlinear interactions in ways that are quite unexpected and even counterintuitive: Contrary to the expectation that two-photon absorption will have no preference for particular frequency components within the spectrum of the pulse, our experiment demonstrates an anomalous, frequency-selective two-photon absorption. The simple model that we use to analyze the experimental data demonstrates the crucial role played by the phase of the optical field. Whereas we take advantage of naturally occurring absorption lines to demonstrate the effect, similar results can be obtained by use of artificial narrow-line absorbers. More generally, similar effects may be expected whenever the phase of the pulse envelope varies abruptly. This property could possibly be used as a diagnostic tool that would detect phase discontinuities in an incident laser pulse. It is also worth noting that the anomalous two-photon absorption that we observe occurs under conditions in which the (classical) linear atmospheric absorption is also anomalous.[24]

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