Ultrafast adiabatic control of reciprocity and coherent backscattering in random scattering media

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ABSTRACT

We experimentally demonstrate ultrafast control over reciprocal light paths in random media. The combination of multiple scattering and coherence of light gives rise to strong interference contributions in light transport. An important interference correction to diffusion theory is the coherent backscattering effect, the constructive interference of reciprocal light paths in the backscattering direction. Our experiments show that the phase coherence between these paths can be suppressed by introducing dynamics faster than the photon dwell time. This adiabatic dephasing is of interest for its potential for controlling weak and strong localization and adiabatic storage and release of photonic information.

Keywords: nanophotonics, multiple scattering, ultrafast, photonic media, nonlinear optics, optical switching

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1. INTRODUCTION

Multiple scattering transport of light in complex photonic media can be considered a model system for quantum transport phenomena. In low-temperature measurements of electronic conductance, it has been observed that quantum-mechanical interference of electron waves scattered by impurities gives rise to new effects on a mesoscopic scale, i.e. between the length scales of individual atoms and the macroscopic system. Constructive interference of time-reversed paths leads to a two times larger probability of an electron wave to return to the same point compared with any other point in space. This phenomenon of weak localization is a precursor of an even stronger localization effect predicted by Anderson, where the wave function itself gets localized by disorder. Light provides exciting opportunities for studying pure wave scattering phenomena like localization [1]. Many more properties can be probed using light waves, such as coherent backscattering, a manifestation of weak localization, and fluctuations and correlations of transmitted light.

The experimental studies of quantum transport in electronic systems is complicated by the presence of many-body interactions which give rise to inelastic processes and dephasing. In comparison, electromagnetic waves interact only weakly and allow investigations of the interplay between multiple scattering and interference. Effects of dephasing can be introduced by adding a nonlinear scattering potential, which effectively takes on the role of many-body interactions. The field of nonlinear transport of light has been actively studied from the theoretical side [2-9]. However experiments in condensed-matter systems have been relatively few [10,11]. We have recently reported ultrafast control over light transport in the strong scattering regime [12,13]. Such effects are of interest for several reasons: i. as a model system for studying quantum interference in the presence of nonlinearity and noise; ii. as a new test for verifying the mesoscopic origin of fluctuations, and iii. for applications in controlling the flow and emission of light in nanophotonic environments.

In recent work, we have shown that ultrafast dephasing is a new and powerful strategy for achieving large nonlinear modulation of light using disordered media. Using only a micrometer thin slab of semiconductor nanowires, ultrafast modulation of individual transmission speckles of up to 50% was demonstrated [12]. The picosecond time scale of this modulation was comparable to the dwell time of light inside the medium. Therefore, it should be possible to control the mode adiabatically, i.e. during the dwell time of light inside the medium. Such adiabatic control has been achieved in

photonic crystal waveguides and microcavities [14-20], but never before in a three-dimensional random medium. As a next step, we showed that we could control the fundamental interference between reciprocal – or time-reversed – light paths [13]. The principle is shown in Fig. 1, where we have drawn an example of light paths travelling from point A to point B inside a random medium. For paths travelling back to the source A, another path can be found where the light follows the exact same microscopic path but in the opposite direction. These reciprocal paths interfere constructively at point A, resulting in a factor of two enhancement of the return probability, and in a concomitant reduction of the diffusion constant. This effect is the principle of weak localization. An associated effect can be observed for waves scattered from random media in reflection. Around the backscattering direction, constructive interference between reciprocal paths gives rise to a peak of enhanced intensity. The height of the peak lies exactly a factor two above the diffuse scattering background (in absence of single scattering), while the angular width of the cone is determined by the product of the wavevector in vacuum and the mean free path of the waves inside the medium, $k_0 l$. The inverse of $k_0 l$ is also known as the photonic strength, as it quantifies the importance of mesoscopic interference corrections in transport.

Excitation with an ultrafast laser beam can result in changes in the scattering potential. If this change is faster than the time the light needs to traverse the multiple scattering path, then the modification of the medium may result in a change of the symmetry between the two reciprocal paths. For weak changes to the medium, the modulation will change mainly the amplitude and/or phase between the two light paths, and dephasing of the two paths will result in a reduced interference contribution. The pulsed laser excitation will have a spatiotemporal profile, which may be used to optimize the dephasing response by adding additional dynamics to the system, or by imprinting a spatially inhomogeneous nonlinear scattering potential.

In our earlier work we explored the presence of reciprocity breaking in GaP random media using the coherent backscattering effect. We showed that the coherent backscattering is reduced under the condition that the pump excited the medium *after* the probe light, but within the photon dwell time. In the current work, we present additional results demonstrating the dependence of the effects on pump power. Also we show that no reciprocity breaking is observed for layers of TiO_2 paint, which is attributed to a difference in the nonlinear response of the TiO_2.

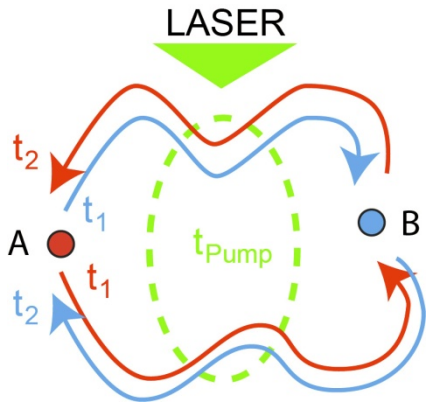

Figure 1 Example of reciprocal light paths travelling from A to B and back to A. The times t_1, t_2 indicate the departure and arrival times of the light. An ultrafast laser pulse is used to break the symmetry of the paths after a time t_{Pump}.

2. METHOD

2.1 Materials

Random photonic materials of sufficient scattering strength are required to observe interference corrections to diffusive transport. In addition, to study effects of nonlinearity, a material with a large nonlinear optical coefficient is desirable. In this studies, we investigated two types of nonlinear scattering media. As in our earlier work, we used porous slabs of gallium phosphide obtained by electrochemical etching of a (100) GaP wafer. The thickness was controlled by the etching time, while porosity was optimized to achieve a moderately strong scattering strength of $k_0 l = 4.5$. While stronger scattering GaP down to $k_0 l \sim 2.5$ can be achieved by increasing the material porosity, it was found that better reciprocity

breaking effects could be obtained in less strongly scattering samples. This may be due to a higher filling fraction of nonlinear semiconductor in the samples of lower porosity, combined to the different ratio between the length scales of scattering and nonlinearity. Also the value of $k_0 l \sim 5$ is better suited for our experimental arrangement as explained in Sec. 2.2.

The second sample under study was a thick (1 mm) slab of TiO_2 powder (Sigma-Aldrich) pressed between two glass cover glasses. The sample was also moderately scattering with an estimated value of $k_0 l = 5.8 \pm 0.7$ as fitted from the coherent backscattering cone of Fig. 2.

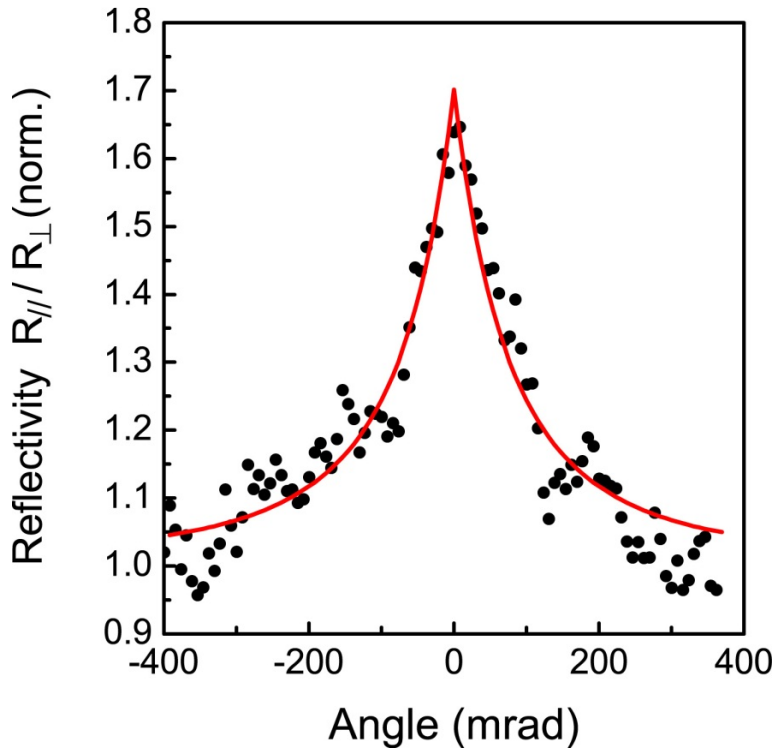

Figure 2. Experimental coherent backscattering cone of TiO_2 slab (dots), with theoretical model fit yielding $k_0 l = 5.8 \pm 0.7$ (line, red).

2.2 Pump-probe coherent backscattering experiment

Pump-probe experiments were performed using a regenerative amplifier setup (Coherent RegA) operating at 250 kHz repetition frequency and with a pulse duration of 0.2 ps. An optical parametric amplifier was used to obtain a probe wavelength of 630 nm. At this wavelength, the samples studied were strongly scattering with negligible absorption. Figure 3 shows a sketch of the experimental setup. The challenge in this experiment is to achieve a sufficient pumping power while still having sufficient angular resolution. In particular, the angular range near the center of the cone contains information on very long light paths involving thousands of scattering events. The power density requirement limited the size of the pump focus to a diameter of about 25 μm, which set the angular resolution by the diffraction limit to about 30 mrad. The total width of the cone which could be measured was defined by the accessible scan range of 5cm to around 700 mrad. The combination of angular range and resolution provided a constraint on the scattering strength of the materials that could be meaningfully studied of around $k_0 l \sim 5$.

The principle of the experiment is shown in the lower half of Fig. 3. The pump pulse photoexcites the free carriers in the medium. The combination of absorption and dephasing process changes the reflectivity. Generally absorption lowers the diffuse albedo by several percent. The dephasing results in a suppression of the coherent backscattering effect through (partial) breaking of reciprocity and absorption. This contribution is found in the parallel polarization channel and mainly for angles close to the backscattering angle corresponding to long light paths. Effectively, more energy is removed from the cone than from the diffuse background, giving rise to a feature in the differential reflectivity.

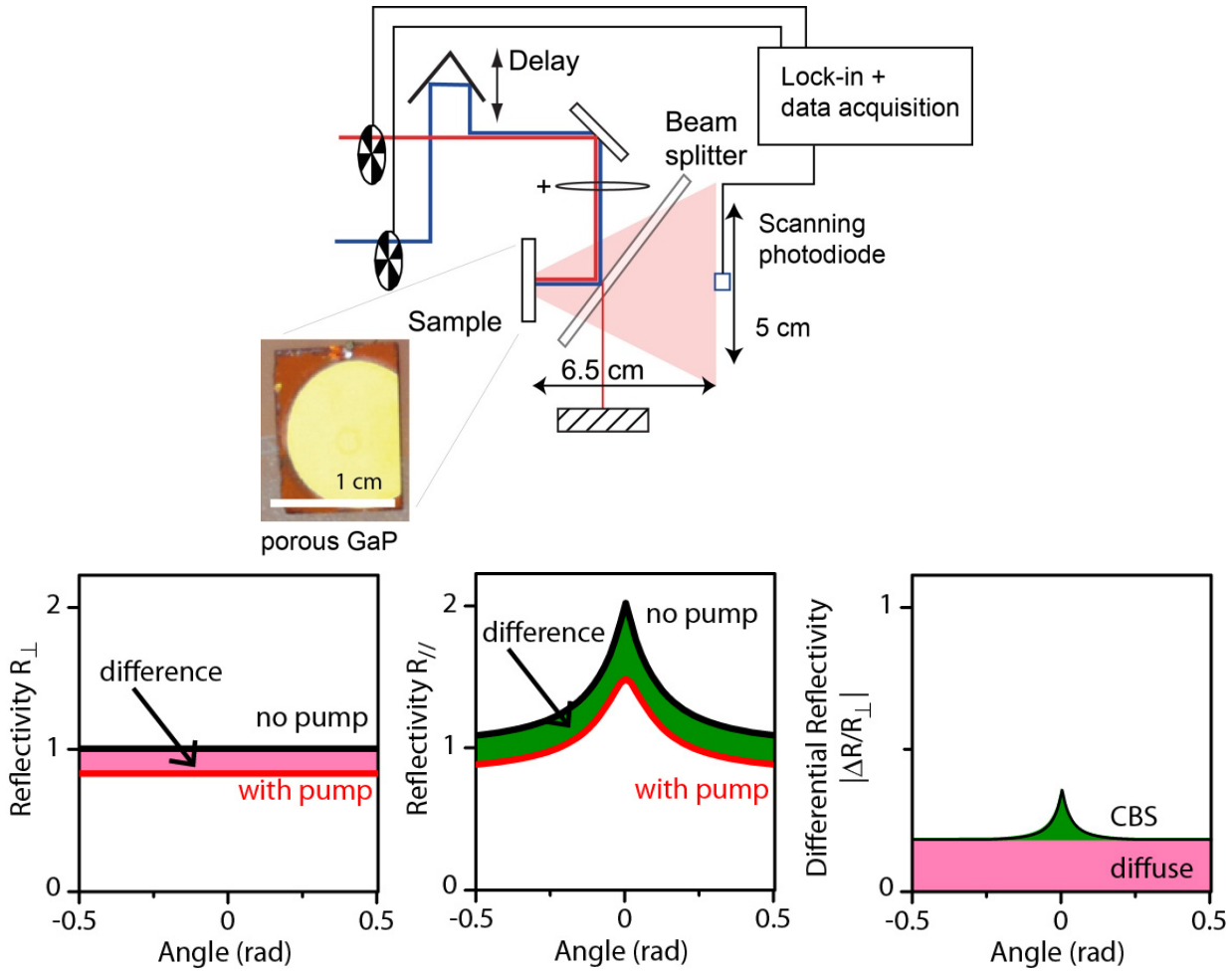

Figure 3. (Top) Experimental setup for ultrafast pump-probe coherent backscattering spectroscopy. Inset shows porous GaP sample under study. (Bottom) Explanation of principle of experiment. Pump lowers reflectivity of diffuse intensity $R_\perp$. For coherent backscattering the same reduction is found, plus an additional effect in the center of the cone due to nonlinear dephasing (reciprocity breaking) of long light paths. In the differential reflectivity $\Delta R/R_\perp$ this is observed as a peak in the normalized signal.

3. EXPERIMENT

3.1 Power dependence of the nonlinear response and reciprocity breaking in a porous GaP slab

The first demonstration of ultrafast reciprocity breaking was presented in Ref. [13]. It was demonstrated that ultrafast dephasing provided a reciprocity breaking under the condition that probe light entered the medium before the arrival of the pump pulse. Here, we present the dependence of this effect on the excitation power.

Figure 4 (left) shows the dependence of the pump-probe reflectivity signal in the diffuse scattering channel. The pump-probe intensity before zero delay time shows a tail, corresponding to arrival of probe light before the excitation by the pump. The shape of this tail can be explained by the path length distribution of light in the medium, longer light paths remaining for a longer time and thus being susceptible to pump excitation for large negative pump-probe delay times [13]. The right-hand panel of Fig. 4 shows the comparison between the pump-probe transient reflectivity for the diffuse

(cross-polarized) and the coherent backscattering (parallel-polarized) channels. The coherent backscattering shows a markedly different shape of the tail, which results from the additional contribution of reciprocity breaking [13].

To quantify this difference, we took the ratio of the parallel to cross-polarized pump-probe signals as a measure of the additional nonlinear modulation caused by reciprocity breaking. This ration should be less than 2.0 for effects caused by absorption only, as absorption similarly modifies the diffuse and coherent backscattering albedo by removing light paths. In practice, this ratio is smaller because of the reduced experimental enhancement factor of the CBS intensity. Figure 5 shows this ratio for characteristic delay times of -1.5 ps and +1.5 ps respectively. At 1.5 ps delay, the additional effects of reciprocity breaking are absent and the nonlinear signal ratio amounts to 1.72. At -1.5 ps, the ratio is increased to 2.6 because of additional nonlinear reciprocity breaking of long light paths in the CBS intensity. The accuracy is limited, especially at low fluences, by the signal to noise ratio of our setup (colored dots indicate two different datasets).

In order to model the dependence on excitation power, we used Monte Carlo numerical simulations of the nonlinear coherent-backscattering cone such as explained in Ref. [13]. In GaP, the complex refractive index change is governed by the free-carrier nonlinearity. As a first approximation, we assume both the real and the imaginary parts of Δn to scale equally with pump power starting from the fitted value of $\Delta n = 1.4 \times 10^{-2} + 1.4 \times 10^{-3}$ i for maximum pump power [13]. Absorption is governed by Lambert-Beer's law which predicts an exponential decay of intensity over the path length. An expansion for small absorption results in an approximately linear dependence on Δn_{Im}. Dephasing however depends on the accumulated phase shift $\mathrm{Re}[\exp(i\Delta n_{Re} kL)]$, which for small Δn_{Re} scales quadratically with power. Therefore, for very low pump fluences, the dephasing term becomes negligible and the nonlinear CBS enhancement in Fig. 5 approaches the limiting value - with only absorption - of 1.92 (this value is less than 2 as the model accounts for a non-perfect angular resolution of the CBS cone). For very large pump fluences, the absorption coefficient becomes so large that the nonlinear cone approaches the linear CBS cone; the nonlinear pump probe enhancement flattens off at 10 mJ/cm^2 and eventually drops down to the value for absorption only for very high pump power. Our experimental data does not allow to draw conclusions on the correctness of this model. On average, experiments indicate a smaller dependence on pump power than expected from linear scaling of both absorption and dephasing. Possible differences could be given by variations in spatiotemporal dynamics as a function of pump fluence, as spatiotemporal effects were included in the model [13]. Additionally, contributions of nonlinear (two-photon) absorption could cause a stronger dependence of the photoinduced absorption on pump intensity. Future studies will have to explore these contributions into more detail.

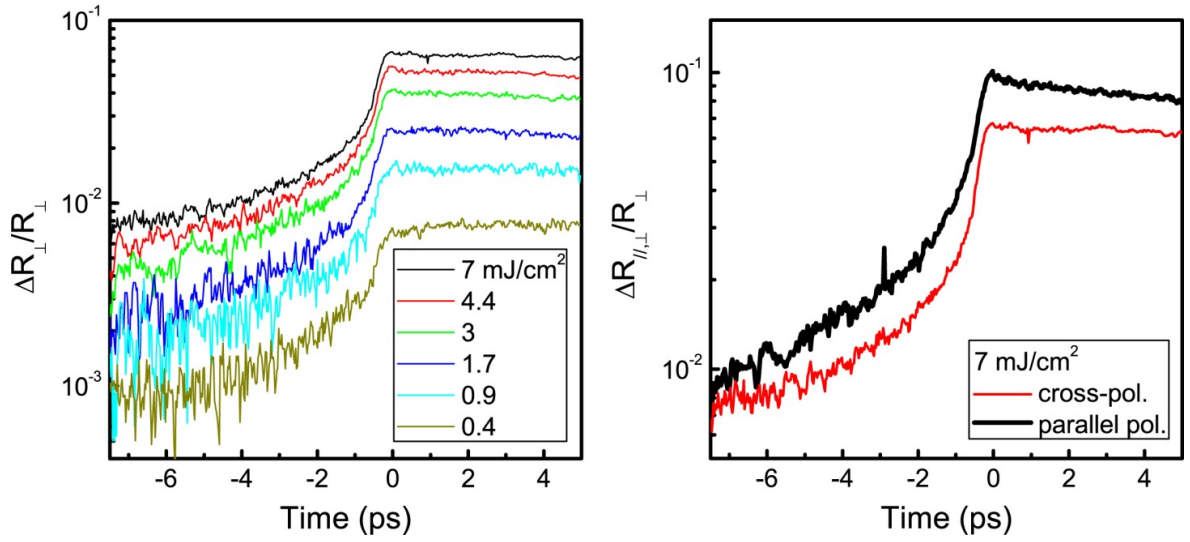

Figure 4 (left) Time dependent transient reflectivity of porous GaP layer for cross-polarized polarization (diffuse component). Negative times correspond to the pump pulse arriving after the probe. Different curves were taken at different values of pump fluence as indicated in the legend. (right) Time-dependent reflectivity $\Delta R / R_\perp$ at highest pump fluence, for diffuse background $\Delta R_\perp$ (cross polarized) and center of the coherent backscattering cone $\Delta R_{//}$ (parallel polarized).

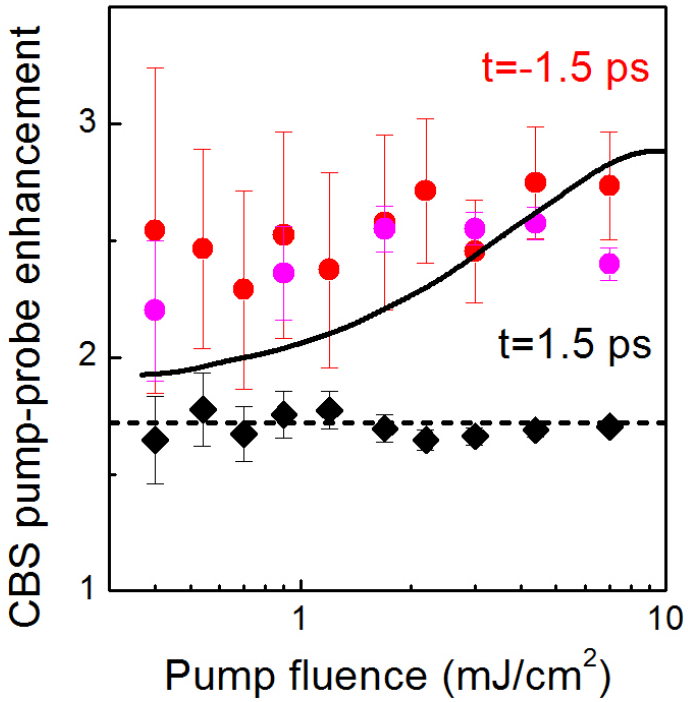

Figure 5 Ratio of the parallel polarized to cross-polarized pump-probe reflectivity ("CBS pump-probe enhancement") for delay times of -1.5 ps (red/purple dots) and +1.5 ps (black diamonds), versus pump fluence. Curve represents Monte Carlo simulation.

3.2 Absence of reciprocity breaking in TiO_2 powder

Our results on porous GaP indicated pronounced contributions from nonlinear reciprocity breaking. It is of interest to find out if similar results can be obtained for other types of scattering materials. For this purpose, we investigated the nonlinear response of the TiO_2 powder sample using exactly the same configuration. Figure 2 shows the tail of the diffuse reflectivity at negative pump-probe delay times. Similarly to the GaP slab, we find a strong transient absorption signal of up to 10% and a pronounced tail at negative delay times, with a time constant given by leakage of light out of the excited region in the transverse direction (Muskens2012). These results show that pump-probe transient absorption can be used as a generic tool for characterizing path length distributions in different types of random media.

We subsequently measured the reflectivity in the peak of the CBS cone to obtain the nonlinear pump-probe enhancement ratio in the same way as Figs. 4 and 5, at the highest pump fluence of 7 mJ/cm^2. Figure 7 shows a comparison between the ratio versus time for the TiO_2 sample (red circles) and the GaP slab (green triangles). Remarkably, no effect of reciprocity breaking could be observed for the TiO_2 layer. Instead the enhancement ratio decays slowly from an initial value of around 1.8. The reduction of the enhancement at long negative times is caused by a convolution of the narrowing nonlinear CBS cone with the instrumental angular resolution of 30 mrad.

The combination of a large transient absorption signal with no reciprocity breaking indicates that, under the condition of ultrafast pumping at 400 nm, the nonlinear response of TiO_2 around 630 nm is governed by the imaginary part Δn_{Im}. The excitation wavelength of 400 nm is in the tail of the band-to-band transition TiO_2. The transient absorption signals of nanostructured TiO_2 under these excitation conditions are given by the dynamics of free and surface trapped electrons [21]. For III-V semiconductors like GaP, the ultrafast response is governed by the free-carrier nonlinearity for above-bandgap excitation [22]. In addition, the direct electronic bandgap of GaP is located in the proximity of the probe wavelength and effects of transient bleaching and bandgap renormalization may contribute in its vicinity. While more systematic investigations are required, our preliminary results show that the choice of material plays a crucial role in the observability of dephasing and reciprocity breaking effects in random media.

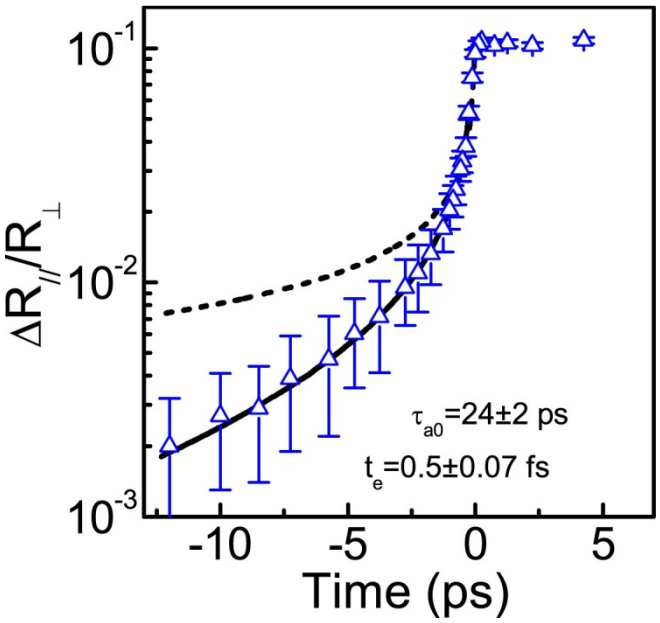

Figure 6 Time dependent reflectivity of TiO_2 layer for cross-polarized polarization (diffuse component). Negative times correspond to the pump pulse arriving after the probe, i.e. light is already inside the medium when it is switched. The tail was analyzed using a convolution of the path length distribution with the material response function (dashed) and including the finite excitation geometry (solid line) with an exponential leakage of light outside the region over a time constant of 24 ps. Fitted extinction time t_e=0.5 fs.

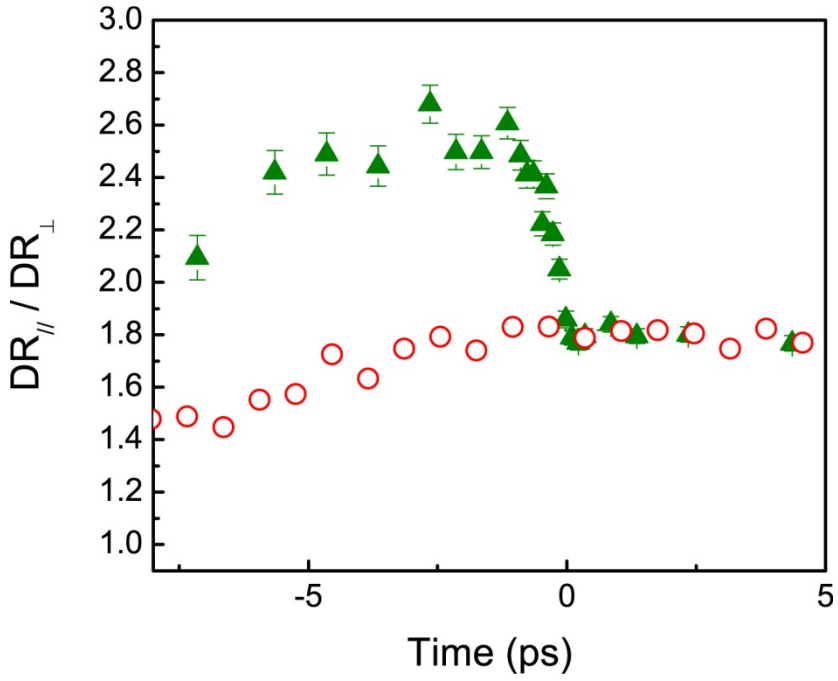

Figure 7. Time-dependence of the CBS pump-probe enhancement ratio $DR_{//}/DR_{\perp}$ for the porous GaP sample (green triangles) and the TiO_2 powder sample. (black line, red circles indicate 10-point moving average).

4. CONCLUSIONS

In conclusion, we have investigated effects of reciprocity breaking in GaP and TiO_2 random media. Our earlier results, demonstrating partial reciprocity breaking in a porous GaP layer, were extended to include the dependence on excitation power. For TiO_2 we found a large contribution of transient absorption, however no signature of reciprocity breaking was found. This result shows that the choice of material is crucial for observing these new and remarkable effects in random media. Future work may optimize the wavelength range of study and the choice of material to enhance even more these effects. In addition, it will be of interest to investigate other adiabatic modulation effects such as frequency conversion, adiabatic storage and release, and eventually nonlinear Anderson localization.

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