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Detectivity enhancement in THz electrooptical sampling

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We demonstrate and discuss a simple scheme that significantly enhances the detectivity of THz electro-optical sampling by introducing a sequence of Brewster windows that increases the ellipticity of the probe beam. By varying the window material or the number of Brewster windows, the enhancement factor can be adjusted; we demonstrate an enhancement factor of ≈ 20 with four ZnSe Brewster windows. The scheme is particularly useful when very small THz fields are to be measured in connection with low-repetition rate amplified Ti:S laser systems.

I. INTRODUCTION

In recent years, there has been a tremendous development in single cycle THz pulses produced from femtosecond lasers [1–4] with applications for example in imaging [5–7], in the non-destructive detection of explosives [8], as well as in other fields of spectroscopy [9, 10]. In time-domain THz spectroscopy, one often uses the generated single cycle THz pulses only as bright light source with coherent detection, but one does not make explicit use of the fact that it is also a short pulse. Optical pump-THz-probe experiments, on the other hand, do so and have been applied to study e.g. phonon dynamics [11, 12], ultrafast carrier dynamics in various materials [13–19], other excitations in strongly correlated materials [20, 21], as well as low frequency vibrational dynamics in molecular liquids [10, 22, 23]. Most time-resolved THz setups use light from a laser oscillator for THz generation and detection, where the high repetition rate and high stability of the laser helps tremendously to achieve very good signal-to-noise ratios. But as the field moves towards nonlinear THz experiments and optical-pump-THz-probe experiments, higher laser powers are needed that call for amplified laser systems with much lower repetition rates. At the same time, the emitted THz fields originating from a nonlinear interaction might be very weak. On the other hand, too much of single pulse energy is available from amplified laser systems as probing light for electro-optical sampling that would saturate any detector. Here, we describe a scheme that makes use of the extra amount of single pulse energy and thereby enhances the THz detectivity by a significant factor ≈ 20 .

II. RESULTS

We used a typical optical-pump-THz-probe experimental setup [24] based on an amplified Ti:S laser system, but for the purpose of this discussion, only the THz-detection part is relevant, which used a 0.1 mm GaP [110] crystal for electro-optic sampling (see Fig. 1). A THz field at the detection crystal renders it birefringent with an optical axis 45° to the laser polarization. An initially *s*-polarized 800 nm probe beam will thus become elliptically polarized with a small *p*-polarization compo-

nent. In a standard electro-optic sampling scheme [4], the light would then be passed through a quarter wave plate, which results in a distorted circular polarized beam, and then through a Wollaston polarizer, which separates *p*- and *s*-polarization components whose difference is measured by two balanced photo diodes. The *p*-polarization component after the detection crystal is proportional to the THz-field and the rotation sense of the elliptically polarized reflects the sign of the THz field, but the ellipticity can be very small, depending on the strength of the THz field. In order to reduce the large *s*-polarization component, the beam first passed through a set of four Brewster windows before the Wollaston polarizer in our implementation (see Fig. 1). These Brewster windows (ideally) transmit 100% of the *p*-polarization component, but reflect a certain fraction of the *s*-polarization component, thus increasing the ellipticity of the beam and, in a relative sense, the *p*-polarization component.

The Brewster windows have a small wedge of 1° to avoid that multiply reflected beams hit the detector. This wedge causes a deflection $d\theta$ of the beam of $d\theta \approx (n^2 - 1)\alpha \approx 5^\circ$ (where α is wedge angle, and n the index of refraction). Since it is critical for the overall performance of the setup that every ZnSe surface is hit at the Brewster angle as closely as possible, the tilt of second and third Brewster window is corrected for that deflection angle (see Fig. 1). The overall transmission through the four Brewster window setup was measured to be $t=0.24\%$, which perfectly matches the expected value for the index of refraction of $n=2.52$ for ZnSe at 800 nm with a Brewster angle of 68° .

Fig. 2 shows the THz pulses measured with (red line) and without (blue line) Brewster windows by scanning the 800 nm probe pulse relative to the THz pulse. In these two experiments, the amount of light at the two photodiodes of the balanced detection was kept the same with the help of additional attenuators in front of the balanced detection, which compensated the effect of the Brewster windows in the measurement without them. Comparing both measurements, we obtain an enhancement factor of ≈ 18 (Fig. 2, blue dots).

FIG. 1: Electro-optical sampling setup with four Brewster windows in order to increase the ellipticity of the probe beam. The bottom row shows the polarization states at various positions along the beam path.

III. DISCUSSION

To describe the polarization states at the various positions in the setup, we use the Jones matrix formalism. We start from linearly s -polarized probe light:

$$p_{in} = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \quad (1)$$

The detection crystal acts as a phase retarder with small retardation ϕ , that is proportional to the THz field:

$$C = \frac{1}{2} \begin{pmatrix} e^{i\phi} + e^{-i\phi} & e^{i\phi} - e^{-i\phi} \\ e^{i\phi} - e^{-i\phi} & e^{i\phi} + e^{-i\phi} \end{pmatrix}. \quad (2)$$

The Brewster windows reduce the s -polarization component by a field attenuation factor a while it fully transmits the p -polarization component:

$$B = \begin{pmatrix} a & 0 \\ 0 & 1 \end{pmatrix}, \quad (3)$$

and the quarter-wave plate is described as:

$$Q = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & i \\ i & 1 \end{pmatrix}. \quad (4)$$

The final polarization state thus becomes:

$$p = Q.B.C.p_{in} = \frac{1}{\sqrt{2}} \begin{pmatrix} a \cos \phi - \sin \phi \\ i(a \cos \phi + \sin \phi) \end{pmatrix} \quad (5)$$

The balance detection measures the difference in intensity of the s - and p -polarization components, which we normalize to the total amount of light on the two detectors:

$$S = \frac{|p_s|^2 - |p_p|^2}{|p_s|^2 + |p_p|^2} \approx \frac{2\phi}{a} \quad (6)$$

FIG. 2: THz pulses measured with (red) and without (blue) Brewster windows. The amount of light at the two photodiodes of the balanced detection was the same in both measurements. The blue line shows the raw data without Brewster windows, the blue dots the data up-scaled by a factor 18.

where the last step is valid for $\phi \ll a$. We see that the relative signal is enhanced by a factor $1/a = 1/\sqrt{t}$ (where t is the corresponding intensity attenuation factor). For the concrete situation in our experiment, we would expect an enhancement factor of ≈ 20 ; slightly higher than the experimental value of ≈ 18 . We attribute the small deviation from the theoretical value to the fact that we do not hit all surfaces of the Brewster windows with perfectly the Brewster angle due small to misalignments and the wedges of the Brewster windows.

With the choice of the window material and the number of Brewster windows, one can adjust the enhancement factor. Four ZnSe windows turned out to be a good compromise between enhancement factor on the one

hand, and the available probe pulse energies in connection with the saturation limit of the photo diodes on the other hand. The maximum enhancement factor is also limited by the condition $\phi \ll a$ (Eq. 6); if a is set too small, the signal no longer scales linearly with THz-field. In the limit of $a = 0$, when only the p -polarization component is transmitted, one could in fact no longer distinguish the signs of the THz field and only the absolute-square of the THz field would be detected.

One may think of the sequence of Brewster windows as a polarizer with an extinction ratio of $1 : 2.4 \cdot 10^{-3}$. Before implementing these Brewster windows, we tested a combination of two film-polarizers with a comparable extinction ratio of $1 : 10^{-3}$. However, the enhancement factor was only ≈ 6 , significantly smaller than the theo-

retical value $1/\sqrt{t}$, presumably since the small amount of transmitted s -polarization is no longer in a clean polarization state afterwards.

In conclusion, we have introduced and discussed a simple scheme that significantly enhances the detectivity of THz electro-optical sampling. The scheme is particularly useful when very small THz fields are to be measured in connection with low-repetition rate amplified Ti:S laser systems. For example, the 2D-Raman-THz experiments of Ref. [24] would not have been possible without that enhancement of the THz detectivity.

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- [1] B. Ferguson and X. C. Zhang, *Nat. Mate.* **1**, 26 (2002).
 - [2] K. Reimann, *Rep. Prog. Phys.* **70**, 1597 (2007).
 - [3] J. Faure, J. van Tilborg, R. A. Kaindl, and W. P. Lee-mans, *Optical and Quantum Electronics* **36**, 681 (2004).
 - [4] Y.-S. Lee, *Principles of Terahertz Science and Technology* (Springer, 2010).
 - [5] E. Pickwell and V. P. Wallace., *J. Phys. D: Appl. Phys.* **02** (2009).
 - [6] D. M. Mittleman, R. H. Jacobsen, and M. C. Nuss, *IEEE J. Sel. Top. Quantum Electron* **2**(3), 679 (1996).
 - [7] K. J. Siebert, T. Löffler, H. Quast, M. Thomson, T. Bauer, R. Leonhardt, S. Czausch, and H. G. Roskos, *Physics in Medicine and Biology* **47**, 3743 (2002).
 - [8] H. Zhong, A. Redo-Sanchez, and X.-C. Zhang, *Int. J. High Speed Electron. Syst.* **17**, 239 (2007).
 - [9] M. Theuer, S. S. Harsha, D. Molter, G. Torosyan, and R. Beigang, *Chemphyschem* **12**, 2695 (2011).
 - [10] C. A. Schmuttenmaer, *Chem. Rev.* **104**, 1759 (2004).
 - [11] P. U. Jepsen, W. Schairer, I. H. Libon, U. Lemmer, N. Hecker, M. Birkholz, K. Lips, and M. Schall, *App. Phys. Lett.* **79** (2001).
 - [12] T. Kampfrath, L. Perfetti, F. Schapper, C. Frischkorn, and M. Wolf, *Phys. Rev. Lett.* **95**, 187403 (2005).
 - [13] J. Zielbauer and M. Wegener, *Appl. Phys. Lett.* **68**, 1223 (1996).
 - [14] M. C. Nuss, D. H. Auston, and F. Capasso, *Phys. Rev. Lett.* **58**, 2355 (1987).
 - [15] B. N. Flanders, D. C. Arnett, and N. F. Scherer, *IEEE J. Sel. Top. Quantum Electron* **4**, 353 (1998).
 - [16] H. Némec, K. Kadlec, and P. Kužel, *J. Chem. Phys.* **117**, 8454 (2002).
 - [17] E. Knoesel, M. Bonn, J. Shan, F. Wang, and T. F. Heinz, *J. Chem. Phys.* **121**, 394 (2004).
 - [18] P. A. George, J. Strait, J. Dawlaty, S. Shivaraman, M. Chandrashekar, F. Rana, and M. G. Spencer, *Nano Letters* **8**, 4248 (2008).
 - [19] M. Breusing, C. Ropers, and T. Elsaesser, *Phys. Rev. Lett.* **102**, 086809 (2009).
 - [20] D. Polli, M. Rini, S. Wall, R. W. Schoenlein, Y. Tomioka, Y. Tokura, G. Cerullo, and A. Cavalleri, *Nature Materials* **6**, 643 (2007).
 - [21] K. W. Kim, A. Pashkin, H. Schaefer, M. Beyer, M. Porer, T. Wolf, C. Bernhard, J. Demsar, R. Huber, and A. Leitenstorfer, *Nature Materials* **11**, 497 (2012).
 - [22] G. Haran, W. D. Sun, K. Wynne, and R. M. Hochstrasser, *Chem. Phys. Lett.* **274**, 365 (1997).
 - [23] R. McElroy and K. Wynne, *Phys. Rev. Lett.* **79**, 3078 (1997).
 - [24] J. Savolainen, S. Ahmed, and P. Hamm, *Proc. Natl. Acad. Sci. USA* **110**, 11725 (2013).