

EDITORIAL • OPEN ACCESS

## Focus on nonlinear terahertz studies

To cite this article: Alfred Leitenstorfer *et al* 2014 *New J. Phys.* **16** 045016

View the [article online](#) for updates and enhancements.

### Related content

- [Terahertz studies of 2D and 3D topological transitions](#)
- [Table-top sources of ultrashort THz pulses](#)
- [Intense ultrashort terahertz pulses: generation and applications](#)

### Recent citations

- [Single Free-Falling Droplet of Liquid Metal as a Source of Directional Terahertz Radiation](#)  
Petr M. Solyankin *et al*
- [Terahertz-driven ultrafast recovery of plasmon resonance in photoexcited nanoantennas on GaAs](#)  
Ali Mousavian *et al*
- [A terahertz-driven non-equilibrium phase transition in a room temperature atomic vapour](#)  
C. G. Wade *et al*

## Editorial

# Focus on nonlinear terahertz studies

**Alfred Leitenstorfer<sup>1</sup>, Keith A Nelson<sup>2</sup>, Klaus Reimann<sup>3</sup> and Koichiro Tanaka<sup>4</sup>**

<sup>1</sup>Department of Physics, Center for Applied Photonics, University of Konstanz, 78457 Konstanz, Germany

<sup>2</sup>Department of Chemistry, Massachusetts Institute of Technology, Cambridge, MA 02139, USA

<sup>3</sup>Max-Born-Institut für Nichtlineare Optik and Kurzzeitspektroskopie, 12489 Berlin, Germany

<sup>4</sup>Department of Physics, Kyoto University, Sakyo-ku, Kyoto 606-8502, Japan

E-mail: [reimann@mbi-berlin.de](mailto:reimann@mbi-berlin.de)

Received 25 February 2014

Accepted for publication 25 February 2014

Published 22 April 2014

*New Journal of Physics* **16** (2014) 045016

doi:[10.1088/1367-2630/16/4/045016](https://doi.org/10.1088/1367-2630/16/4/045016)

### Abstract

Resulting from the availability of improved sources, research in the terahertz (THz) spectral range has increased dramatically over the last decade, leading essentially to the disappearance of the so-called ‘THz gap’. While most work to date has been carried out with THz radiation of low field amplitude, a growing number of experiments are using THz radiation with large electric and magnetic fields that induce nonlinearities in the system under study. This ‘focus on’ collection contains a number of articles, both experimental and theoretical, in the new subfield of THz nonlinear optics and spectroscopy on various systems, among them molecular gases, superconductors, semiconductors, antiferromagnets and graphene.

Keywords: terahertz radiation, light–matter interaction, Rabi oscillations, high-field transport

A nonlinear response to the electromagnetic field requires high intensity. At first glance, it may seem improbable to achieve sufficiently large intensities (or electric field amplitudes) in the THz



Content from this work may be used under the terms of the [Creative Commons Attribution 3.0 licence](https://creativecommons.org/licenses/by/3.0/). Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.

frequency range. Since intensity is pulse energy divided by pulse length and illuminated area, in the THz range all three factors reduce the reachable intensity: the pulse energy is much smaller than at higher optical frequencies, the pulses are longer (the minimum pulse length can be taken roughly as the inverse of the frequency), and the illuminated area is at least on the order of the wavelength squared. As the papers in this ‘focus on’ collection show, despite the lower intensities in the THz range, it is possible to observe THz nonlinearities. What are the reasons for this fact? For an answer to this question one first has to consider when nonlinearities occur. One possibility also present in other spectral ranges is a transition driven resonantly with high field amplitude. Here a nonlinearity occurs if the population transfer from the ground to the excited state is appreciable. With the transition dipole moment  $d$  and the electric field  $E$  this is equivalent to  $dE \ll \hbar\nu$ , i.e. if the Rabi frequency reaches the order of magnitude of the transition frequency. Here it helps that in the THz range the transition dipole moments are typically larger than for higher frequencies. This can be seen simply from the uncertainty relation: increasing the size of a system reduces the momentum uncertainty and thus the energy, resulting eventually in THz-frequency transitions. At the same time, transition dipole moments scale with the system dimensions. Another possibility unique to the THz range that can lead to nonlinearities is the acceleration of charged carriers in the THz field. Here nonlinearities occur if the kinetic energy gained from the accelerating field becomes comparable to energies in the system, e.g. to the band gap in a semiconductor. Since the kinetic energy gained is proportional to the time the field acts in one direction, THz pulses have the advantage of comparatively long acceleration times. Therefore, in the THz range, much lower intensities are required to observe nonlinearities than for higher frequencies. Even these intensities only became available in the last decade or so, rendering the study of THz nonlinearities a relatively new field.

The generation of high THz intensities is still a topic of research. In this collection, several papers consider THz generation in a laser-generated plasma [1–3]. These sources can achieve very broad spectra, extending up to a frequency of  $\approx 100$  THz (corresponding to a wavelength of  $3 \mu\text{m}$ ) if very short pump pulses are used. For many applications, they have the disadvantage that the spatial profile of their emission is quite complex. Three further papers consider other methods for THz generation, one by intracavity mixing [4] and two with quantum cascade lasers [5, 6].

Apart from generation, detection is also an important topic. In [7], the authors present a scheme to allow for the determination of the square of the electric field as a function of time over a broad frequency range by four-wave mixing in gases.

The remaining papers consider the action of high-intensity THz pulses on solids, among them antiferromagnets, superconductors, graphene and semiconductors.

The paper on antiferromagnets [8] is unique in this collection of articles in that the relevant interaction is with the THz magnetic field, while in all other papers it is only with the THz electric field. In this respect, one should note that an electromagnetic wave with an electric field amplitude of  $3 \text{ MV cm}^{-1}$  is accompanied by a magnetic field with an amplitude of 1 T.

A natural object of THz studies is superconductors, since in conventional superconductors the superconducting gap is in the THz range. In this collection, there are three papers on superconductors, two on conventional [9, 10] and one on high-temperature [11] superconductors.

In graphene, which can be considered a two-dimensional semiconductor with a zero band gap, the THz range is particularly interesting, since here THz pulses can induce both resonant transitions between the valence and conduction band (one can find points in the  $k$  space where

the energy difference is equal to the THz frequency), and lead to electrical transport by moving the charge carriers in real and reciprocal space. This interplay between inter- and intraband transitions is calculated in [12]. The predicted [12, 13] generation of THz higher harmonics was not observed in [14], but in [15]. In [16] surface plasmons in graphene are considered.

The majority of papers in this collection are concerned with the study of semiconductors [8, 17–23]. While their band gaps are typically much larger than THz photon energies (an exception to this is the narrow-gap semiconductor InSb [8]), one can still have resonant transitions in the THz range, either between discrete levels of impurities [17] or between discrete states caused by spatial confinement, e.g. in quantum wells [18–20]. As mentioned in the introduction, a second way for nonlinearities to occur is via transport over a large part of the Brillouin zone [21, 22].

This ‘focus on’ collection shows that it is possible to perform nonlinear THz studies for a wide range of materials. In many cases, such studies lead to new results not available with other techniques. Further advances in the generation and detection of THz pulses and in understanding their interaction with matter will certainly lead to a continued series of exciting results in the future.

## References

- [1] Oh T I, You Y S, Jhajj N, Rosenthal E W, Milchberg H M and Kim K Y 2013 Intense terahertz generation in two-color laser filamentation: energy scaling with terawatt laser systems *New J. Phys.* **15** 075002
- [2] Klarskov P, Strikwerda A C, Iwaszczuk K and Jepsen P U 2013 Experimental three-dimensional beam profiling and modeling of a terahertz beam generated from a two-color air plasma *New J. Phys.* **15** 075012
- [3] Blank V, Thomson M D and Roskos H G 2013 Spatio-spectral characteristics of ultra-broadband THz emission from two-colour photoexcited gas plasmas and their impact for nonlinear spectroscopy *New J. Phys.* **15** 075023
- [4] Kiessling J, Breunig I, Schunemann P G, Buse K and Vodopyanov K L 2013 High power and spectral purity continuous-wave photonic THz source tunable from 1 to 4.5 THz for nonlinear molecular spectroscopy *New J. Phys.* **15** 105014
- [5] Wang F, Guo X G, Wang C and Cao J C 2013 Ultrafast population dynamics in electrically modulated terahertz quantum cascade lasers *New J. Phys.* **15** 075009
- [6] Iotti R C and Rossi F 2013 Coupled carrier–phonon nonequilibrium dynamics in terahertz quantum cascade lasers: a Monte Carlo analysis *New J. Phys.* **15** 075027
- [7] Clerici M, Faccio D, Caspani L, Peccianti M, Yaakobi O, Schmidt B E, Shalaby M, Vidal F, Légaré F and Ozaki T 2013 Spectrally resolved wave-mixing between near- and far-infrared pulses in gas *New J. Phys.* **15** 125011
- [8] Pashkin A, Sell A, Kampfrath T and Huber R 2013 Electric and magnetic terahertz nonlinearities resolved on the sub-cycle scale *New J. Phys.* **15** 065003
- [9] Zachmann M, Croitoru M D, Vagov A, Axt V M, Papenkort T and Kuhn T 2013 Ultrafast terahertz-field-induced dynamics of superconducting bulk and quasi-1D samples *New J. Phys.* **15** 055016
- [10] Zhang C *et al* 2013 Nonlinear response of superconducting NbN thin film and NbN metamaterial induced by intense terahertz pulses *New J. Phys.* **15** 055017
- [11] Grady N K *et al* 2013 Nonlinear high-temperature superconducting terahertz metamaterials *New J. Phys.* **15** 105016
- [12] Ishikawa K L 2013 Electronic response of graphene to an ultrashort intense terahertz radiation pulse *New J. Phys.* **15** 055021

- [13] Ishikawa K L 2010 Nonlinear optical response of graphene in time domain *Phys. Rev. B* **82** 201402(R)
- [14] Paul M J, Chang Y C, Thompson Z J, Stickel A, Wardini J, Choi H, Minot E D, Norris T B and Lee Y 2013 High-field terahertz response of graphene *New J. Phys.* **15** 085019
- [15] Bowlan P, Martinez-Moreno E, Reimann K, Elsaesser T and Woerner M 2014 Ultrafast terahertz response of multi-layer graphene in the nonperturbative regime *Phys. Rev. B* **89** 041408(R)
- [16] Watanabe T, Fukushima T, Yabe Y, Tombet S A B, Satou A, Dubinov A A, Aleshkin V Y, Mitin V, Ryzhii V and Otsuji T 2013 The gain enhancement effect of surface plasmon polaritons on terahertz stimulated emission in optically pumped monolayer graphene *New J. Phys.* **15** 075003
- [17] Nagai M, Kamon Y, Minowa Y, Matsubara E and Ashida M 2013 Coherent transitions between the shallow acceptor levels in germanium using intense THz pulses *New J. Phys.* **15** 065012
- [18] Teich M, Wagner M, Schneider H and Helm M 2013 Semiconductor quantum well excitons in strong, narrowband terahertz fields *New J. Phys.* **15** 065007
- [19] Dietze D, Darmo J and Unterrainer K 2013 Efficient population transfer in modulation doped single quantum wells by intense few-cycle terahertz pulses *New J. Phys.* **15** 065014
- [20] Köster N S, Klettke A C, Ewers B, Woscholski R, Cecchi S, Chrastina D, Isella G, Kira M, Koch S W and Chatterjee S 2013 Controlling the polarization dynamics by strong THz fields in photoexcited germanium quantum wells *New J. Phys.* **15** 075004
- [21] Xie X T, Zhu B F and Liu R B 2013 Effects of excitation frequency on high-order terahertz sideband generation in semiconductors *New J. Phys.* **15** 105015
- [22] Yang F and Liu R B 2013 Berry phases of quantum trajectories of optically excited electron–hole pairs in semiconductors under strong terahertz fields *New J. Phys.* **15** 115005
- [23] Hase M, Katsuragawa M, Constantinescu A M and Petek H 2013 Coherent phonon-induced optical modulation in semiconductors at terahertz frequencies *New J. Phys.* **15** 055018