

TERA-MIR Radiation: Materials, Generation, Detection And Applications

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Abstract

The main objective of MPNS COST ACTION MP1204 is to advance novel materials, concepts and device designs for generating and detecting THz (0.3 THz to 10 THz) and Mid Infrared (10 THz to 100 THz) radiation using semiconductor, superconductor, metamaterials and lasers. This special edition summarizes part of the progress achieved in the first year of our network by covering a wide range of related topics, e.g. fundamental studies of intervalence THz polaritons and entangled excitons to the design and development of THz and MIR Quantum Cascade Lasers and novel antennas, filters and metamaterials. Spectroscopy and modelling of the interaction of THz radiation with biomatter are investigated. Mid infrared detection and emission are modelled and developed including supercontinuum studies. Relevant results for nitrides, bismides and graphene are discussed covering industrial and academic applications, theory and experiments, illustrating the impact of a synergistic approach to THz and MIR within our TERA-MIR concept.

Key words: TERA-MIR; Midinfrared; Terahertz, Sources; Detectors; Materials; Polaritons; Antipolaritons.

Many substances exhibit rotational and vibrational transitions in the TERA-MIR region (0.3 THz to 100 THz) hence giving access to a spectroscopic analysis of a large variety of molecules which play a key role in security as well as various other areas, e.g. air pollution, climate research, industrial process control, agriculture, food industry, workplace safety and medical diagnostics can be monitored by sensing and identifying them via TERA-MIR absorption “finger prints” [1]. Most plastics, textiles and paper are nearly transparent for THz radiation. Notably CBRN (Chemical, biological, radiological and nuclear) agents, explosives, illegal drugs can be detected by their characteristic absorption spectra at THz frequencies with high selectivity and resolution in applications fields as industrial quality inspection control, customs inspection and security screening. Moreover, THz and MIR radiation has no endangering effects on human beings and enables higher contrast for “soft matter” than x-rays [2]. In comparison to standard optical technologies for wavelengths up to about 2 μ m, sources and detectors for THz and MIR have not yet reached this level of maturity and there is still a large gap for features like wavelength tunability, spectral purity, high power and room temperature operation, which all are necessary for commercial applications.

Plastic or ceramics are detected of by X-rays very poorly especially against a background of human body. Unlike X-rays, THz (or T-wave) is not a dangerous radiation, and in some cases T-wave sensors can reveal not only the shape of a hidden object but also its chemical composition. This unique combination of traits make T-waves perfect for effective applications like explosive detection, and security applications. Besides, T-rays have high resolution in 3D space in case of THz ultra-short pulses. The possibility to analyse chemical composition of substances by spectroscopic methods is of big interest. Even in case if the substance is in the plastic tank or under the cloth. However, there are many open problems on the path to practical and routine use of THz.

The main THz and MIR source is the quantum cascade laser (QCL). This special edition covers QCL development as well as a number of other alternatives and new materials and devices consistently summarized efforts to turn advanced technologies into affordable commercial devices throughout the TERA-MIR spectral range and exploit their enormous potential for security applications. Starting with QCLs,

M. Szymański et al., discuss metal–metal waveguides for terahertz quantum-cascade lasers have and modelled as planar, multilayer structures. The optical properties of constituent materials were calculated according to Drude–Lorenz model. They concentrate on selecting the proper metallic material for claddings to minimize the waveguide losses. In addition, they analyse the consequences of inserting Ti separation layers between claddings and semiconductor core for blocking the destructive diffusion of metals into the active layer and improving the adhesion. Further experimental details can be found in Ref. [3].

Emilia Pruszyńska-Karbownik et al, present intra-pulse measurements of a QCL beam have been reported. The laser had an active region made of $\text{Al}_{0.45}\text{Ga}_{0.55}\text{As}/\text{GaAs}$ coupled quantum wells and emitted mid-infrared radiation ($\lambda \sim 9.4 \mu\text{m}$). The experiment was made by using a goniometric profilometer with an oscilloscope triggered with a supply, which made the intra-pulse measurement possible. The beam-steering phenomenon was observed with the maximal beam deflection of 17° . The experimental results are explained by a two-mode theoretical model. The near field of the laser radiation is calculated according to the effective index method and transposed to the far field numerically according to Huygens principle.

P. Karbownik et al, develops chip bonding technology of QCLs. Various solders were investigated and compared in terms of their thermal resistance and induced stress. The particular attention has been paid to Au–Au die bonding, which seems to be a promising alternative to the choice between hard and soft solder bonding of QCLs operating from cryogenic temperatures up to room temperatures. High performance room temperature operation of GaAs/AlGaAs QCLs has been achieved with the state-of-the-art parameters. This type of bonding can push mid-IR and THz emitters toward higher working temperatures.

F. Janiak shows that testing structures imitating the operational laser device might be performed by spectroscopy experiments realized within the Fourier Transformed Infrared Spectroscopy based concept. [4]. AlGaAs/GaAs superlattices designed as an active and injector region of QCL were investigated. The band structure of the formed conduction mini bands were studied and the applied growth rates investigated by the Stokes shift between the

emission and absorption spectra. These optical properties is important for designing and construction QCL and other low dimensional semiconductor lasers emitting in a mid and far infrared spectral range.

Other types of heterostructures and design techniques have also been investigated:

Jelena Radovanovic et al., discuss a method which delivers optimal cubic GaN/AlGaN quantum well (QW) profiles such that both the Stark effect and peak intersubband absorption from the ground to the first excited electronic state, in a prescribed range of bias electric fields, are maximized has been proposed. The method relies on the Genetic Algorithm which finds globally optimal structures with a predefined number of embedded layers. Simple rectangular quantum wells with embedded step layers for applications in tunable mid-infrared photodetectors have been studied, including the effects of band nonparabolicity.

Other functionalities for the THz range have been investigated. Compared to the visible and infrared regions of the electromagnetic spectrum, development of optical components in the terahertz region has progressed slowly even though the feature size of structures makes many of them easily attainable by many conventional manufacturing techniques. Of these, the performance of components with machined metal surfaces, such as filters, typically has suffered from the errors and inconsistencies in the manufacturing which has led many to manufacture these using more expensive deposition and processing tools, thus

L. Juul et al deliver an efficient numerical method to design terahertz photomixers. The simulations were benchmarked using measured power levels from results published in the literature. The method has been applied to two new photomixer designs based on the high impedance T-match antenna with bias supply DC-blocking structures for either a uniplanar layout or a multilayer structure for improved device reliability. Manufacturability is favoured by avoiding the use of airbridges, substrate thinning or under-etching. The estimated output power of the improved design is 9.0 μW , which is an improvement of 3 times over the reference photomixer.

I. Faragai and M.F. Pereira, further bridge the gap between basic physics and applications, by discussing intra-valence-band THz polaritons, which have strong potential for device applications and are a challenging field of fundamental studies are discussed with mathematical model based on a nonlinear dielectric constant approach to the optical susceptibility. The coupling of TE-polarized THz radiation with an intervalence band transition in GaAs/Al_{0.3}Ga_{0.7}As multiple quantum wells embedded in microcavities has been analysed. The energy dispersions relations leading to THz polaritons are investigated. Here we focus on the interplay of cavity length and size of the active region against the dephasing mechanisms on polariton dispersion relations [5].

Bismide and nitride-based semiconductor play a major role in our Network [6].

C. Oriaku and M.F. Pereira deliver an efficient approximation to the complex many body problem of luminescence in semiconductors to the case of mid infrared luminescence of dilute nitrides has been delivered by The results are in very good agreement with recent mid infrared

experimental data from teams in our Network [6]. This theory is obtained after systematic simplifications of a more complete optical response for semiconductor materials, obtained by self-consistent evaluation of Manybody Nonequilibrium Green's Functions (NEGF). Efficient NEGF-based numerical methods used here have been successfully applied to both intersubband [7,8] and interband transitions [9,10] in quantum wells and superlattices. This paper starts from a similar approach that can also describe superlattices as effective 3D anisotropic media [11] and leads to very accurate approximations.

GaAsBi-based structures have a strong potential for applications from the mid- to the near infrared region. It is anticipated that GaAsBi with more than 10 % Bi could be prospective for the active layers of telecom wavelength laser diodes because for that material composition non-radiative losses would be effectively reduced due to the larger than the energy band gap spin orbit splitting in the valence band. The main challenge in this field is the intensity of the radiative transitions that becomes significantly smaller (nearly by four orders of magnitude) in the bismide layers with more than 5 % Bi. Thus,

Renata Butkute et al, show strong photoluminescence for these materials at wavelengths ranging from 1300 to 1500 nm in GaAsBi layers grown by migration-enhanced epitaxy and annealed at higher than 600°C temperatures. The structural investigations revealed that annealing reduces Bi content in GaAsBi lattice by precipitating Bi in the nanometer-scale clusters. It was suggested that enhancement of photoluminescence signal could be caused by the semimetal-semiconductor transition due to quantum confinement effect in Bi clusters. Complementing these investigations,

A. Koroliov et al, report the optical absorption bleaching in several GaAs_{1-x}Bi_x layers. Absorption saturation spectra in GaAs_{1-x}Bi_x samples have been measured by optical pump-and-probe technique, absorption bleaching was observed up to the wavelengths of 1600 nm; its typical recovery times were of the order of several picoseconds. Short carrier lifetimes as well as saturation fluencies comparable to their values in the best known semiconductor saturated absorbers make the investigated layers promising for SESAM applications. We also demonstrated the experimental techniques suitable for carrier lifetime and semiconductor band gap determination by nonlinear absorption measurements.

There is a strong interest in our network for application of graphene and graphene-related phases (polygraphene, multilayered graphene etc.) in FE transistors [12], the use of graphene as a gate electrode in IIIIV based FETs, novel detectors for radiation in MIR/THz region etc. Therefore, the founding of technologies for reliable deposition of graphene and graphene related phases becomes of increasing interest. Thus by the work of

T. Milenov and al., it is established that polygraphene layers could be deposited by sublimations of pyrolytic carbon as show in this edition: the surface modification of different poligraphene and multilayered graphene layers by low-energy Ar⁺ plasma (in the range of 1015 Ar⁺/cm²) even for 8 s significantly worsened the quality of films as well as that the graphene has a pronounced self-healing ability. The thinning of thin pyrolytical carbon films by 1015 Ar⁺/cm² plasma is considered and it is established that few-layered polygraphene can

be obtained by 30 s surface plasma modification and 7 min thermal annealing at 2700C in air atmosphere.

T. Takan et al show that by using a novel, high power Yb:doped pulsed nanosecond fiber laser system with exceptional beam quality, aluminum metal surfaces can be machined with high precision leading to a high quality band pass filter working in the terahertz frequency range. The produced structures were modeled by utilizing the obtained structural parameters in a waveguide configuration and then characterized by the existing home-built time domain terahertz spectrometers at METU, Turkey. The results show a near 100 % power transmission at the desired terahertz frequency range which suggests that these manufacturing techniques can be used to produce low-cost THz filters [13,14].

Finally, another topic of strong interest of our Network is the interaction of radiation and bio matter [15]. Particularly, the nonenzymatic attachment of sugars to proteins, namely glycation, is accelerated under diabetic conditions. Monitoring the glycated human serum albumin (HSA) levels gives the short term variation of glucose concentration in diabetic patients' blood. Therefore, a significant effort was made by

I. Vasile presents measurements of glycated HSA, including by spectroscopic methods such as Raman. Research presented here, used THz spectroscopy to monitor HSA glycation in time (over 5, 7 and 11 weeks). Different sugar types have different reactivity; therefore we also addressed the reducing sugar influence on glycation by performing in vitro HSA glycation by both glucose and fructose. Since residues protonation state influences their susceptibility for glycation, we incubated HSA with sugars at two pH values: 7 and 8. Our results show that THz absorption decreases with the incubation time of HSA with sugars. At the incubation times we considered, the most significant differences were obtained on HSA samples glycated using glucose. Differences between samples glycated by glucose and by fructose show that glycation by glucose is a slower process. At pH 7, glycation by glucose is slower than at pH 8, while glycation by fructose is slightly faster at pH 7 than at pH 8. Glycated HSA models with different degrees of glycation were built by molecular modeling. Simulated THz spectra of the models are in good agreement with the experimental data. All these show that THz spectroscopy could monitor the progression of glycation in time and that it is sensitive to reducing sugars or pH value used in the glycation process.

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References

- [1] B. Ferguson and X.-C. Zhang, "Materials for Terahertz Science and Technology," *Nature Materials* 1, 26-33 (2002).
- [2] For a review see *Terahertz and Mid Infrared Radiation: Detection of Explosives and CBRN (Using Terahertz)*, Edited by Mauro F. Pereira and Oleksiy Shulika, NATO Science for Peace and Security Series - B: Physics and Biophysics, Springer (2014). DOI 10.1007/978-94-017-8572-3; *Terahertz and Mid Infrared Radiation, Generation, Detection and Applications*, NATO Science for Peace and Security Series B: Physics and Biophysics, Springer (2011). Pereira, Mauro F.; Shulika, Oleksiy (Eds.) ISBN: 978-94-007-0768-9.
- [3] Szerling, K. Kosiel, M. Szymański, Z. Wasilewski, K. Gołaszewska, A. Łaszcz, M. Płuska, A. Trajnerowicz, M. Sakowicz, M. Walczakowski, N. Pałka, R. Jakiela, A. Piotrowska, "Processing of AlGaAs/GaAs QC structures for terahertz laser", *Proc. SPIE 9199, Terahertz Emitters, Receivers, and Applications V*, 919903 (September 5, 2014); DOI :10.1117/12.2061997
- [4] Janiak, F., Motyka, M., Sek, G., Dyksik, M., Ryczko, K., Misiewicz, J., Weih, R., Hoefling, S., Kamp, M., Patriarche, G.: Effect of arsenic on the optical properties of GaSb-based type II quantum wells with quaternary GaInAsSb layers. *J. Appl. Phys.* 114(22), 223510 (2013).
- [5] M. F. Pereira Jr. and I.A. Faragai, Coupling of THz radiation with intervalence band transitions in microcavities *Optics Express*, Vol. 22 Issue 3, pp.3439-3446 (2014). DOI:10.1364/OE.22.003439
- [6] M. F. Pereira Jr. and S. Tomić, Intersubband gain without global inversion through dilute nitride band engineering, *Appl. Phys. Lett.* 98, 061101 (2011).
- [7] A Krier, M de la Mare, P J Carrington, M Thompson, Q Zhuang, A Patanè and R Kudrawiec, *Semicond. Sci. Technol.* 27, 094009 (2012)
- [8] M.F. Pereira Jr, R. Nelander, A. Wacker, D.G. Revin, M.R.Soulby, L.R. Wilson, J.W. Cockburn, A.B. Krysa, J.S. Roberts, and R.J. Airey, Characterization of Intersubband Devices Combining a Nonequilibrium Many Body Theory with Transmission Spectroscopy Experiments, *Journal of Materials Science: Materials in Electronics* 18, 689 (2007)
- [9] M.F. Pereira Jr., Microscopic approach for intersubband-based thermophotovoltaic structures in the THz and Mid Infrared, *JOSA B* Vol. 28, Iss. 8, pp. 2014–2017 (2011).
- [10] H. Grempe, A. Diessel, W. Ebeling, J. Gutowski, K. Schull, B. Jobst, D. Hommel, M.F. Pereira and K. Henneberger, High-density effects, stimulated emission, and electrooptical properties of ZnCdSe/ZnSe single quantum wells and laser diodes, *Physica Status Solidi B* 194, 199 (1996)
- [11] M.F. Pereira Jr., "Analytical solutions for the optical absorption of superlattices", *Phys. Rev. B* 52, (1995).
- [12] Graphene field-effect transistors as room-temperature terahertz detectors, L. Vicarelli, M. S. Vitiello, D. Coquillat, A. Lombardo, A. C. Ferrari, W. Knap, M. Polini, V. Pellegrini and A. Tredicucci, *Nature Material* 11, 865–871 (2012). DOI: doi:10.1038/nmat3417

- [13] Z. Ozer, S. Gok , H. Altan, F. Severcan, "Concentration Based Measurement Studies of L-Tryptophan using THz-Time Domain Spectroscopy (THz-TDS)," *Applied Spectroscopy*, 68 (1) 95-100 (2014).
- [14] H. Keskin, H. Altan, S. Yavaş, F. Ö. İlday, K. Eken, A. B. Şahin, "Development of a Rapid-Scan Fiber-Integrated Terahertz Spectrometer," *Optical and Quantum Electronics* 46, 495–503 (2014).
- [15] M. Mernea, O. Calborean, O. Grigore, T. Dascalu, D. F. Mihailescu, " Validation of protein structural models using THz spectroscopy", *Opt Quant Electron* 46, pp. 505 – 514 (2014).