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**RPPR Final Report**  
as of 08-Nov-2018

Agency Code:

Proposal Number: 65552ELCF

**Agreement Number: W911NF-14-1-0343**

**INVESTIGATOR(S):**

**Name:** Xi-Cheng Zhang  
**Email:** xi-cheng.zhang@rochester.edu  
**Phone Number:** 5852750333  
**Principal:** Y

Organization: **University of Rochester**

Address: ORPA, Rochester, NY 146270140

Country: USA

DUNS Number: 041294109

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**Final Report** for Period Beginning 01-Aug-2014 and Ending 31-Jul-2016

**Title:** Symposium on High-Field THz Wave Generation and Nonlinear THz Physics

**Begin Performance Period:** 01-Aug-2014

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**Report Term:** 0-Other

Submitted By: Xi-Cheng Zhang

Email: xi-cheng.zhang@rochester.edu

Phone: (585) 275-0333

**Distribution Statement:** 1-Approved for public release; distribution is unlimited.

**STEM Degrees:**

**STEM Participants:**

**Major Goals:** Our aim is to develop a state-of-the-art, time-resolved bright THz source with at least 10 MV/cm THz peak field strength. This one-order-of-magnitude higher field strength THz source will open the door for many research frontiers in THz field-matter nonlinear interaction.

**Accomplishments:** We made significant progress in the following aspects: (1) Demonstrate broadband THz generation from liquid water. (2) Terahertz Radiation Enhanced Emission of Fluorescence (REEF) from elongated plasmas and microplasmas in the counter-propagating geometry. (3) Measurement of an extremely large nonlinear refractive index of crystalline ZnSe at terahertz frequencies by a modified Z-scan method. (4) Squeezing the fundamental temperature fluctuations of a high-Q microresonator. (5) Demonstrated THz emission from abruptly-autofocusing (AAF) beam induced air plasma (6) Tested THz emission from micro-plasma generated by dual-color laser beam. (7) Third order nonlinearity at the THz range for different materials. (8) Explored THz-REEF in counter-propagation geometry with different fluorescence lines. (9) Detecting THz with microplasma using THz-REEF technique. (10) We have organized Symposium on "High-Field THz Wave Generation and Nonlinear THz Physics".

**Training Opportunities:** Nothing to Report

## RPPR Final Report as of 08-Nov-2018

### Results Dissemination: Papers published in peer reviewed journals (including submitted)

1. Jin Qi, Yiwen E, Kaia Williams, Jianming Dai, X.-C. Zhang, "Observation of Broadband THz Generation from Liquid water", *Appl. Phys. Lett.*, 111(7), 071103, (2017).
2. Xuan Sun, Rui Luo, X.-C. Zhang, Qiang Lin, Squeezing the fundamental temperature fluctuations of a high-Q microresonator, *Physical Review A* 95, (2017), 023822.
3. Xuan Sun, Hanxiao Liang, Rui Luo, Wei C. Jiang, Xi-Cheng Zhang, and Qiang Lin, "Nonlinear optical oscillation dynamics in high-Q lithium niobate microresonators," *Opt. Express* 25, 13504-13516 (2017)
4. A.P. Shkurinov, A.S. Sinko, P.M. Solyankin, A.V. Borodin, M.N. Esaulkov, V.V. Annenkov, I.A. Kotelnikov, I.V. Timofeev, X.-C. Zhang, Impact of the dipole contribution on the terahertz emission of air-based plasma induced by tightly focused femtosecond laser pulses, *Physical Review E* 95, (2017), 043209.
5. Kang Liu, D. G. Papazoglou, A. D. Koulouklidis, S. Tzortzakis, and X.-C. Zhang, "Enhanced terahertz wave emission from air-plasma tailored by abruptly autofocusing beam," *Optica* 3(6) 605, (2016).
6. L.L. Zhang, T. Wu, H. Zhao, C. Zhang, W. Jin, X.-C. Zhang, Enhanced THz-to-IR emission from gas-surrounded metallic nanostructures by femtosecond laser irradiation, *Optics Communications*, 381, (2016), 414-417.
7. Y.A. Kapoyko, A.A. Drozdov, S.A. Kozlov, X.C. Zhang, Evolution of few-cycle pulses in nonlinear dispersive media: Velocity of the center of mass and root-mean-square duration, *Physical Review A* 94 (2016) 9.
8. Fabrizio Buccheri, Kang Liu, X.-C. Zhang, "Terahertz Radiation Enhanced Emission of Fluorescence from Elongated Plasmas and Microplasmas in the Counter-propagating Geometry," *Appl. Phys. Lett.*, submitted.
9. Anton N. Tsyppkin, Sergey E. Putilin, Maksim S. Kulya, et al., Measurement of an extremely large nonlinear refractive index of crystalline ZnSe at terahertz frequencies by a modified Z-scan method, *Optical Express*, submitted.
10. L.L. Zhang, S.J. Zhang, R. Zhang, T. Wu, Y.J. Zhao, C.L. Zhang, and X.-C. Zhang, "Excitation-Wavelength Dependent Terahertz Wave Polarization Control in Laser-Induced Filament," *Optica*, (2017). Submitted
11. Liangliang Zhang, Cunlin Zhang and X.-C. Zhang, "Bi-directional terahertz-to-infrared emission from metal-coated nanostructures upon femtosecond laser irradiation," *Optical Express*, submitted.
12. Klarskov, Pernille, Tarekegne, Abebe T., Iwaszczuk, Krzysztof, Zhang, X. C., & Jepsen, Peter, Amplification of resonant field enhancement by plasmonic lattice coupling in metallic slit arrays, *Scientific Report*, 6 (2016) 37738.
13. Liangliang Zhang, Ji Zhao, Tong Wu, Cunlin Zhang and X.-C. Zhang, "Terahertz-to-infrared emission through laser excitation of surface plasmons in metal films with porous nanostructures," *Optical Express*, 23, 17185 (2015).
14. Kang Liu, Fabrizio Buccheri, X.-C. Zhang, "Investigation of THz science and technology of micro-plasma," *WuLi (Chinese Physics Today)*, August issue (2015).
15. L. Zhang, K. Mu, Y. Zhou, H. Wang, C. Zhang, X.C. Zhang, High-power THz to IR emission by femtosecond laser irradiation of random 2D metallic nanostructures, *Scientific Report*, 5 (2015) 12536.
16. Liangliang Zhang, Kaijun Mu, Ji Zhao, Tong Wu, Hai Wang, Cunlin Zhang, and X.-C. Zhang, "Intense thermal terahertz-to-infrared emission from random metallic nanostructures under femtosecond laser irradiation," *Optical Express* 23, 14211 (2015).
17. Liangliang Zhang, Fabrizio Buccheri, Cunlin Zhang and X.-C. Zhang, "Terahertz emission from thin metal films with porous nanostructures," *Applied Physics Letters* 107, 071107 (2015).
18. Mikhail Esaulkov, Petr Solyankin, Artem Sidorov, Lyubov Parshina, Artem Makarevich, Qi Jin, Qin Luo, Oleg Novodvorsky, Andrey Kaul, Elena Cherepetskaya, Alexander Shkurinov, Vladimir Makarov, Xi Cheng Zhang, "Emission of Terahertz pulses from vanadium dioxide films undergoing metal-insulator phase transition," *Optica* 2 (9), 790-796 (2015).
19. F. Buccheri, X.-C. Zhang, Terahertz emission from laser-induced microplasma in ambient air, *Optica*, 2 (2015) 366-369.
20. K. Dolgaleva, D. V. Materikina, R. W. Boyd, and S. A. Kozlov, "Prediction of an extremely large nonlinear refractive index for crystals at terahertz frequencies", *Phys. Rev. A* 92, 023809 (2015) DOI: 10.1103/PhysRevA.92.023809.
21. Jianming Dai and X.-C. Zhang, "Terahertz wave generation from thin metal films excited by asymmetrical optical fields," *Optics Letters*, 39, 777 (2014).
22. Xiaofei Lu and X.-C. Zhang, "Investigation of ultra-broadband terahertz time-domain spectroscopy with terahertz wave gas photonics," *Frontiers of Optoelectronics*, 7, 121-155 (2014).
23. I-Chen Ho and X.-C. Zhang, "Application of broadband terahertz spectroscopy in semiconductor nonlinear dynamics." *Frontiers of Optoelectronics* 7, 220-242 (2014).
24. Benjamin Clough and X.-C. Zhang, "Toward remote sensing with broadband terahertz waves," *Frontiers of Optoelectronics* 7, 199-219 (2014).
25. Yuting W. Chen and X.-C. Zhang, "Anti-reflection implementations for terahertz waves," *Frontiers of Optoelectronics* 7, 243-262 (2014).

## RPPR Final Report as of 08-Nov-2018

26. Jingle Liu and X.-C. Zhang, "Terahertz radiation-enhanced-emission-of-fluorescence," *Frontiers of Optoelectronics* 7, 156-198 (2014).
27. Benjamin Clough, Jianming Dai, and X.-C. Zhang, "Laser Air Photonics: Covering the Terahertz Gap and Beyond," Review paper, *Chinese Journal of Physics* 52, 416, (2014).
28. Betty Sun and X.-C. Zhang, "Terahertz radiation in alkali vapor plasmas," *Applied Physics Letters* 104, 191106 (2014). doi.org/10.1063/1.4876602.
29. Yan Peng, Xiangqian Chen, Yunyan Zhou, Gongjie Xu, Bin Cai, Yiming Zhu, Jian Xu, Ron Henderson, and Jianming Dai, "Annealing-Insensitive "Black Silicon" with High Infrared Absorption ", *Journal of Applied Physics* 116, 073102 (2014).

### Conference Proceedings

#### a. Presentations at meetings, but not published in conference proceedings

1. "From Speculation to Demonstration: THz Wave Emission from Water," Physics Department, Capital Normal University of Beijing, Beijing, China, June 19, 2017.
2. "Let light shine out of darkness" Princeton International School of Math and Sciences, Princeton, NY, May 12, 2017.
3. "THz: Imaging Beyond Light" Open Readings 2017, 60th International Conference for Students of Physics and Natural Sciences, Vilnius, Lithuania, March 15, 2017.
4. "The Institute of Optics, A jewel in the crown," Academy of Opto-Electronics, Chinese Academy of Science, Beijing, China, Dec. 27, 2016.
5. "The Institute of Optics, Micro-Plasma and Extreme THz Science," Jiliang University, Nov. 8, 2016.
6. "Extreme THz Science," Westlake Photonics Symposium, Zhejiang University, Hangzhou, China, Nov. 7, 2016.
7. "Enhanced terahertz wave emission from air-plasma tailored by abruptly autofocusing laser beams," 8th ISUPTW, Chongqing, China, Oct. 11, 2016
8. "The State of The Institute of Optics," New Graduate students town hall meeting, The Institute of Optics, University of Rochester, September 14, 2016.
9. "Vision" Hong Kong University of Science and Technology, Hong Kong, September 6, 2016.
10. "The Institute of Optics: A Jewel in the Crown," Shandong University of Beijing, Beijing, China, July 18, 2016.
11. "The Institute of Optics: A Jewel in the Crown, Bright THz source and Micro-plasma," Capital Normal University of Beijing, Beijing, China, July 2, 2016.
12. "The Institute of Optics: A Jewel in the Crown," Huazhong University of Science and Technology, Wuhan, China, June 28, 2016.
13. X.-C. Zhang, "Bright THz & Nonlinear THz Field-Matter Interaction," ARO Workshop, University of Rochester, Rochester, USA, June 15, 2016.
14. X.-C. Zhang, "THz wave photonics: bridging the "gap" and beyond," Rochester Perspective Students, Rochester, January 28, 2016.
15. X.-C. Zhang, "Perspectives of Extreme THz Science," International Consortium of THz Photonics and Optoelectronics, Lomonosov Moscow State University, December 16, 2015.
16. X.-C. Zhang, "Passion in THz Science and Technology", Chongqing Instit

## RPPR Final Report as of 08-Nov-2018

**Honors and Awards:** 2016-17 X.-C. Zhang, Scientific Advisor, Capital Normal University of Beijing, China  
2017 Kang Liu, Rochester Precision Optics Award for Outstanding Graduate Projects  
2017 X.-C. Zhang, Australian Academy of Science Selby Fellow, Australia  
2016 Fabrizio Buccheri, Rochester Precision Optics Award for Outstanding Graduate Projects  
2015 X.-C. Zhang, Honorable Professor, Jilin University, China  
2015 X.-C. Zhang, Light of Green and Intelligent Chair Professor at Chongqing Institute of Green X.-C. Zhang, and Intelligent Technology  
2015 X.-C. Zhang, Strategic Advisory Board Member, Chongqing Institute of Green and Intelligent Technology, CAS, China  
2015 Kang Liu, Best Student Paper Award 1st Place, 40th International Conference on Infrared Millimeter and Terahertz Waves (IRMMW-THz 2015), Hong Kong, August 27, (2015)  
2015 Robert Boyd, IEEE Photonics Society Distinguished Traveling Lecturer, 2015.  
2015 Xi-Cheng Zhang, Scientific Advisor, National Research University of Information Technology, Mechanics and Optics (ITMO), St. Petersburg, Russia  
2015 Xi-Cheng Zhang, Kenneth J. Button Prize, International Society of Infrared, Millimeter, and Terahertz Waves.  
2014 Robert Boyd, Elevated to Fellow Status, SPIE 2014  
2014 Robert Boyd, Quantum Electronics Award, IEEE Photonics Society  
2014 Robert Boyd, Honorary Doctorate, University of Glasgow

### **Protocol Activity Status:**

**Technology Transfer:** Nothing to Report

### **PARTICIPANTS:**

**Participant Type:** PD/PI

**Participant:** Xicheng Zhang

**Person Months Worked:** 1.00

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

**Funding Support:**

**THz Workshop on Bright THz source and nonlinear field-matter interaction**  
**Final Progress Report**

Report Period: 08/01/2016--07/31/2017

Grant Contract #: W911NF-14-1-0356

Director, and M. Parker Givens Professor  
The Institute of Optics  
University of Rochester  
275 Hutchison Rd., 114 Wilmot Building  
Rochester, NY, 14627-0186 USA  
Phone: (585) 275-0333  
Fax: (585) 273-1072  
zhangxc@rochester.edu

Submit to

Dr. Joe X. Qiu  
Program Manager  
U.S. Army Research Office  
RDRL-ROE-L  
4300 S. Miami Blvd.  
Durham, NC 27703-9142  
Joe.x.qiu.civ@mail.mil  
1-919-549-4297

Abstract:

We have organized Symposium on “bright terahertz (THz) sources and nonlinear THz field-matter interaction” from the Army Research Office. This symposium provided a forum for exchanging information on generation, detection and applications of intense pulsed THz waves. The symposium brought experts together to discuss recent research findings, and provided an assessment of current and future of bright THz source and nonlinear interaction with matters. This two-days symposium was held on June 2016 in Rochester, NY.

The bottleneck for exploring new cutting edge research and broader applications, following the significant development of THz science and technology in the late 80's, is the lack of bright, high performance, and cost effective THz sources. After significant development of pulsed THz science and technology in the late 80's, the bottlenecks for exploring new cutting edge research and boarder applications are the lack of bright, portable, high performance, and cost effective THz sources. It is important to make assessment for state-of-the-art, time-resolved tabletop bright THz source.

The bright THz source is essential for exploring nonlinear THz field-matter interaction and spectroscopy. The record of THz field strength is about 1 MV/cm or lower in laser-induced gas plasma with two-color laser excitation in dry nitrogen. With our proposed THz source driven by longer-wavelength pulses, THz field strength greater than 10 MV/cm is feasible. This bright THz source will open the door for many research topics in nonlinear THz field-matter interaction.

The THz frequency range is the last segment of the electromagnetic spectrum to be conquered. Far from being fully exploited, yet it offers great opportunities in science, innovation, new technology and applied fields. Our proposed workshop will provide the needed assessment.

The bottleneck for exploring new cutting edge research and broader applications, following the significant development of THz science and technology in the late 80's, is the lack of bright, high performance, and cost effective THz sources. The world record of pulsed THz field strength from tabletop systems is on the order of 1 MV/cm, which is not sufficient to explore many nonlinear field-matter interaction phenomena. The two current technical approaches for achieving such field strengths utilize nonlinear generation in either laser-induced gas plasma with two-color laser excitation or LiNbO3 with tilted pulse-front optical excitation. We propose to develop a state-of-the-art, time-resolved bright THz source with at least 10 MV/cm THz peak field strength. This one-order-of-magnitude higher field strength THz source will open the door for many research frontiers in THz field-matter nonlinear interaction.

A strong THz field, especially with amplitude in the range between 1 to 10 MV/cm, serves as an essential tool for exploring new THz nonlinear phenomena. For example, at 1 MV/cm, intense THz field-matter interactions including resonant and non-resonant controlled molecular vibration, rotation, and spin precession can be achieved. However, certain physical phenomena may require higher field strength for detailed study. Table 1 lists characteristic energies (as well as frequencies) of processes under study in extreme nonlinear science, including ponderomotive force, Rabi oscillation, tunneling frequency, Bloch oscillation, and Cyclotron resonance. The table shows that THz waves (0.25 and 2.5 THz) require much less field strength than visible or UV sources for such nonlinear interactions. Therefore, our proposed THz source serves as an ideal tool to study these frontiers of basic sciences.

**Table 1.** List of characteristic energies (as well as in frequencies) under study in extreme nonlinear science.

Energy scale	Relation to field	1 meV (0.25 THz)	10 meV (2.5 THz)	1 eV (250 THz)	100 eV
Ponderomotive	$U_p = e^2 E_{THz}^2 / (4m\omega^2)$	3 kV/cm	10 kV/cm	100 kV/cm	1 MV/cm
Rabi ( $d=1e\text{\AA}$ )	$U_R = E_{THz} \cdot d$	100 kV/cm	1 MV/cm	100 MV/cm	-
Tunneling freq	$\Omega_T = e / (\hbar (2mE_b)^{1/2})$	2.6 kV/cm	26 kV/cm	2.6 MV/cm	-
Bloch freq	$\Omega_b = eaE_{THz} / \hbar$	20 kV/cm	200 kV/cm	2 MV/cm	-
Cyclotron freq	$\Omega_c = e / (mc)$	4.2 MV/cm	42 MV/cm	-	-

Table 2 lists peak THz electric fields obtained from different THz sources, including superconductor, Gunn diode THz wave emitter, THz gas laser, quantum cascade laser (QCL), large-aperture electro-optic (LA-EO) crystal, large-aperture photoconductive antenna (LA-PC), free electron laser, APS at Argonne National Laboratory, LiNbO3 crystal with tilted pulse front excitation, THz wave air photonics, and SLAC National Accelerator Laboratory. Free electron laser and other accelerator sources offer both high peak power and average power, but the systems are not tabletop. However, this table might be out of date. Our symposium will provide updated info.

**Table 2.** List of the peak THz electric field strengths from different sources.

THz sources	0~10 V/cm	1~10 kV/cm	10~100 kV/cm	0.1 ~1 MV/cm	1~10 MV/cm
Superconductor <sup>[1]</sup>	█				
Gunn Diode <sup>[2]</sup>	█				
THz Gas Laser <sup>[3]</sup>	█				
QCL <sup>[4]</sup>	█				
LA-EO <sup>[5]</sup>			█		
LA-PC <sup>[6]</sup>			█	█	
Free Electron Laser <sup>[7]</sup>					█
APS					█
LiNbO <sub>3</sub> <sup>[8]</sup>					█

The suggested Bright THz workshop will be two days with a single session. We will invite the most active researchers in THz community.

We invited scientists and researchers from Army, Navy, and Air Force laboratories to attend this Workshop. Some of them were very active in a previous NATO THz Exploratory Team (ET) and a Focus Group (FG) while I served as the Chairman.

List of the top players on high-field THz wave generation and nonlinear THz spectroscopy  
USA Domestic

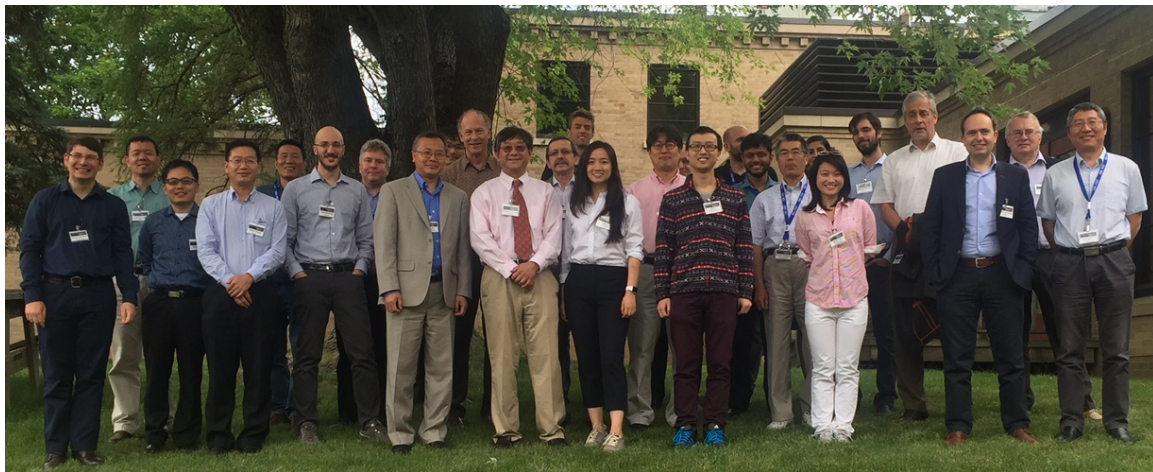
1. G. L. Carr, Brookhaven National Laboratory
2. Aaron Lindenberg, Stanford University or Alan Fisher, SLAC National Accelerator Laboratory (I do not know him well. He is a very good collaborator with Aaron Lindenberg in SLAC)
3. Martin Richardson, University of Central Florida
4. Richard D. Averitt, Boston University (He first reported THz nonlinear responses from metamaterials in collaboration with Keith Nelson)
5. Keith Nelson, MIT
6. Franz X. Kärtner, MIT and University of Hamburg, Germany
7. Haidan Wen, Argonne National Laboratory
8. K-Y Kim, University of Maryland, College Park
9. Howard M. Milchberg, University of Maryland, College Park
10. Gwyn Williams, Jefferson Laboratory
11. Poul B. Petersen (THz wave generation from filaments but not high-field), Cornell University, USA, Email: pbp33@cornell.edu
12. Alex Gaeta, Cornell University, alg3@cornell.edu
13. David A. Reis, Stanford University and SLAC, collaborating with Aaron Lindenberg, Email: dreis@stanford.edu
14. Masashi Yamaguchi, Rensselaer Polytechnic Institute, yamagm@rpi.edu

International

1. Koichiro Tanaka kochan@icems.kyoto-u.ac.jp, Japan
2. H. Hirori, Japan (what work he did? He is the person who is working with Tanaka for >1.0 MV/cm THz electric field generation from LiNbO<sub>3</sub> and can be an alternative invited speaker for the workshop, Email:hirori@icems.kyoto-u.ac.jp)
3. Masa Tonouchi <tonouchi@ile.osaka-u.ac.jp>, Japan (strong field? He is not working in high-field THz sources but probably can be one alternative invited speakers for the workshop)
4. Masaaki Ashida, Japan (what work he did? He or his group demonstrated a bandwidth of over 200 THz, Email: ashida@mp.es.osaka-u.ac.jp)
5. Yasuo Minami or Tohru Suemoto, Japan (1.4 MV/cm from plasma) (email address? Email:minamiyashuo@ynu.ac.jp )
6. János Hebling <hebling@fizika.ttk.pte.hu>, Hungary
7. K. Reimann, M. Woerner, or T. Elsaesser, Germany (email address?Emails: woerner@mbi-berlin.de, reimann@mbi-berlin.de)
8. Hartmut Roskos <roskos@physik.uni-frankfurt.de>, Germany
9. Matthias C. Hoffmann, Germany (He intensively works on nonlinear THz responses or spectroscopy (I couldn't find his email address), we can alternatively contact A. Cavalleri at Email: andrea.cavalleri@mpsd.cfel.de)
10. Alfred Leitenstorfer <aleitens@uni-konstanz.de>, Germany (University of Konstanz)
11. Rupert Huber, Germany (University of Konstanz) (email address?Email: rupert.huber@physik.uni-regensburg.de)

12. Matteo Clerici, Tsuneyuki Ozaki, or Roberto Morandotti, Canada (wavelength scaling of THz radiation) John Ozaki <ozaki@emt.inrs.ca>
13. Peter Uhd Jepsen <puje@fotonik.dtu.dk>, Demark
14. Stelios Tzortzakis stzortz@iesl.forth.gr
15. Frank Hegmann hegmann@phys.ualberta.ca
16. David Cooke <cooke@physics.mcgill.ca>

A two-day ARO THz workshop on “Bright THz Source and Nonlinear THz Field-Matter Interaction” was organized in Rochester, NY, USA in June 2016. The invitation only workshop was a global event where 17 leading researchers from 8 countries gave presentations to share their views, achievements and plans about the future direction of research on intense THz sources, shown in **Fig. 1**.



**Fig.1:** 17 invited speakers from 8 countries presented their work during two days ARO THz workshop on Intense THz Sources - Nonlinear THz field – Matter Interaction at Rochester in June, 2016. Dr. Joe Qiu, ARO Program Manager, sponsored and attended the workshop.