戦略的国際共同研究プログラム(SICORP)　日本－イタリア・ハンガリー共同研究
終了報告書　概要

１．研究課題名：「半導体・金属複合ナノ構造のフェムト秒レーザーパルス照射によるプラズモン増強テラヘルツ波発生（FemtoTera）」

２．研究期間：2014 年 10 月～2017 年 3 月

３．主な参加研究者名：

日本側チーム

	氏名	役職	所属	研究分担
研究代表者	迫田和彰	上席研究員	物質・材料研究機構	試料設計
主たる共同研究者	黒田隆	グループリーダー	物質・材料研究機構	特性評価
主たる共同研究者	間野高明	主幹研究員	物質・材料研究機構	自己成長法の考案
研究期間中の全参加研究者数　　3　名				

相手側チーム

	氏名	役職	所属	研究分担
研究代表者	Stefano Sanguinetti	Professor	University of Milano Bicocca	試料創製と全体の統括
主たる共同研究者	Akos Nemcsics	Managing Researcher	Hungarian Academy of Sciences	キャラクタリゼーション
主たる共同研究者	Caterina Vozzi	Researcher	Institute for Photonics and Nanotechnologies	THz波発生の解析
研究参加者	Cristian Manzoni	Researcher	Institute for Photonics and Nanotechnologies	THz波発生の解析
研究参加者	Lajos Toth	Researcher	Hungarian Academy of Sciences	試料形状の高分解能測定
研究参加者	Bela Pecz	Researcher	Hungarian Academy of Sciences	試料形状の高分解能測定
研究期間中の全参加研究者数　　7　名				

４．共同研究の概要

テラヘルツ（THz）波は化学分析や生体イメージング，非破壊検査，保安検査など多様な分野への応用が期待されている。本研究は，Λ型の電子遷移を実現する量子ドット分子（QDM）と Ga 液滴による金属微小球（MNP）の複合体を作製し，MNP による電磁共鳴効果を利用することで，2 つの電子遷移の干渉による THz 波の高効率発生を目的とする。

５．共同研究の成果

５－１　共同研究の学術成果

この目的のために，申請者らが開発してきた液滴エピタキシー法による QDM と Ga 液滴の作製条件を最適化することで，QDM の 2 つの QD の中央に MNP を作製することに成功した。顕微 PL（フォトルミネッセンス）法で QDM の発光スペクトルを観測

して，THz 領域のエネルギー差をもつ複数の電子遷移の存在を確認した。また，時間分解反射率測定で，QDM と MNP 間の強い相互作用も確認できた。

５－２　国際連携による相乗効果

試料設計，試料作製，形態観察，顕微分光について分担して研究を進めることで，短期間で所期の試料作製に成功した。

５－３　共同研究成果から期待される波及効果

今回の共同研究を通じて，従来，半導体ナノ構造の創製にだけ適用可能であった液滴エピタキシー法が，半導体・金属複合ナノ構造の創製にも拡張できた。特に，複合構造を最適化する作製条件を見出したことは，将来の発展につながる成果である。

Strategic International Collaborative Research Program (SICORP)
Japan－Italy, Hungary Joint Research Program
Executive Summary of Final Report

１．Project Title：「Plasmon-enhanced Tera-Hertz emission by Femtosecond laser pulses of nanostructured semiconductor/metal surfaces (FemtoTera)」

２．Project Period：Oct 1, 2014 ～ Mar 31, 2017

３．Main Participants：

Japan-side（up to 6 people including Principal Investigator）

	Name	Title	Affiliation	Role
PI	Kazuaki Sakoda	Managing Researcher	National Institute for Materials Science	Sample design
Co-PI	Takashi Kuroda	Group Leader	National Institute for Materials Science	Characterization
Co-PI	Takaaki Mano	Principal Researcher	National Institute for Materials Science	Self-assembly
Total number of participating researchers in the project: 3				

Partner-side（up to 6 people including Principal Investigator）

	Name	Title	Affiliation	Role
PI	Stefano Sanguinetti	Professor	University of Milano Bicocca	Sample fabrication
Co-PI	Akos Nemcsics	Managing Researcher	Hungarian Academy of Sciences	Characterization
Co-PI	Caterina Vozzi	Researcher	Institute of Photonics and Nanotechnologies	THz generation
Collaborator	Cristian Manzoni	Researcher	Institute of Photonics and Nanotechnologies	THz generation
Collaborator	Lajos Toth	Researcher	Hungarian Academy of Sciences	High-resolution measurements
Collaborator	Bela Pecz	Researcher	Hungarian Academy of Sciences	High-resolution measurements
Total number of participating researchers in the project: 7				

４．Scope of the joint project

FemtoTera addresses the fundamental technological field of ultrashort pulsed Terahertz (THz) radiation generation for time domain spectroscopy. This is an ever-growing field, due to the important properties of such radiation and its significant applications. One of the more effective way to generate short THz radiation pulses is via femtosecond near-infrared (NIR) laser irradiated on semiconductor/metal surfaces. FemtoTera project targets the study of such THz generation process at surfaces by stimulating with ultrashort laser pulses a semiconductor surface nano-engineered with self-assembled quantum molecules (QDM) - metal nanoparticle (MNP) systems.

The surface is nanostructured with QDM-MNP complexes via bottom-up processes, using the innovative fabrication schemes (Droplet Epitaxy) based on advanced Molecular Beam Epitaxy procedures for the self-assembly and self-alignment of QDM and MNP. Droplet Epitaxy offers the unique opportunity to realize both QDM and MNP within the same bottom-up approach and platform, thus allowing the fabrication of large functionalized surfaces for efficient THz generation.

The interaction effects between ultrashort NIR laser pulses and the nanostructured surface has been investigated, both theoretically and experimentally using the complementary expertise of the EU and Japan groups increasing our understanding of the complex, multi time scale physical interactions between light and matter and their related pho-

to-responsive processes. Relevant effects of the interaction between QNP and ultrashort NIR laser pulses mediated by the MNP have been observed.

The FemtoTera consortium has defined and characterized in detail the QDM-MNP system to meet THz generation requirements by means of experimental and theoretical approaches. The bottom-up nano-structuring of semiconductor surface with QDM-MNP coupled systems, matching the requirements, in terms of quantum confined state energy and localization within the QDM and spatial matching of MNP with the corresponding MNP, was obtained. Accurate morphological and optical characterization has permitted to understand in detail the QDM-MNP growth process and their physical properties in view of their use for efficient THz generation.

５．Outcomes of the joint project

５－１ Intellectual Merit

Design of QDM-MNP systems for THz generation: The project fist task was to design a QDM-MNP structure that reflects the theoretical requirements. For this purpose, EMA electronic state calculations, based on realistic QDM models, taken from AFM measurements of a test sample, have been performed. The relevant parameters were deduced, namely electron and holes wavefunction energies and spatial extension within a single QDM, as a function of the QDM shape and size. These calculations set the size and shape characteristics needed to construct the QDM.

Fabrication and Characterization of QDM-MNP systems for THz generation: Once the growth targets are defined in the design step described previously, through the detailed analysis of the observed dependencies of the QDM morphologies and Ga droplet position on the growth parameters during MBE growth, we could determine the optimum conditions to drive the system towards the targeted QDM electronic states.

５－２ Synergy from the Collaboration

The possibility to make a joint Japanese-European project has permitted the integration of a well-established scientific activities of characterization on nanostructured surfaces in Japan with the growth, structural and optical characterization EU groups working in droplet epitaxy nanostructures and Thz generation by femtosecond laser pulses. This has permitted to integrate a new working practice with a distributed approach, where inputs coming from groups working in different fields and different nations are integrated in a single research line, with frequent interchange of data and discussions about investigation paths and the definition of the optimal design of the nanostructured QDM-MNP hybrid surface.

The consortium successfully coordinated an efficient and interdisciplinary collaboration between the consortium partners to ensure the ideal chain of the comprehension of the mechanisms at the basis of THz generation via nanostructured semiconductor/metal surfaces is achieved.

The network generated by the Femtotera project will continue to collaborate after the project end to the further development of the Femtotera knowledge and technology.

５－３ Potential Impacts on Society

The main impact of the project has been to strongly interlink the four groups participating to the project. This allowed the mutual contamination of expertise between the members and the completion of the complex Femtotera project which required a vast, interdisciplinary knwoledge. In this respect an innovative method for the site matching of QDM and MNP has been developed by the Femtotera consortium which was allowed by the sharing of the scientific competencies of the EU and Japan groups. The scientific results are relevant, clearly opening the way to the use of nanostructured surfaces for the efficient generation of THz radiation via fs laser pulses. The scientific findings will be the argument of two papers in preparation.

別紙２_共同研究における研究成果リスト（迫田・Sanguinetti・Nemcsics 課題）

１．論文発表等

[1] Neul Ha, Xiangming Liu, Takaaki Mano, Takashi Kuroda, Kazutaka Mitsuishi, Andrea Castellano, Stefano Sanguinetti, Takeshi Noda, Yoshiki Sakuma, and Kazuaki Sakoda, “Droplet epitaxial growth of highly symmetric quantum dots emitting at telecommunication wavelengths on InP (111)A”, Applied Physics Letters, vol. 104, no. 14, 143106/p.1-p.4, 2014
[2] G. Sallen, S. Kunz, T. Amand, L. Bouet, T. Kuroda, T. Mano, D. Paget, O. Krebs, X. Marie, K. Sakoda, and B. Urbaszek, “Nuclear magnetization in gallium arsenide quantum dots at zero magnetic field”, Nature Communications, Vol. 5, 3268/p.1-p.7, 2014
[3] Shi-Qiang Li, Peijun Guo, D. Bruce Buchholz, Wei Zhou, Yi Hua, Teri W. Odom, J. B. Ketterson, Leonidas E. Ocola, Kazuaki Sakoda, and Robert P. H. Chang, “Plasmonic-photonic mode coupling in indium-tin-oxide nanorod arrays”, ACS Photonics, Vol. 1, No. 3, p.163-p.172, 2014
[4] Shi-Qiang Li, Wei Zhou, D. Bruce Buchholz, John B. Ketterson, Leonidas E. Ocola, Kazuaki Sakoda, Robert P. H. Chang, “Ultra-sharp plasmonic resonances from monopole optical nanoantenna phased arrays”, Applied Physics Letters, vol. 103, no. 23, 231101/p.1-p.5, 2014
[5] L. Bouet, M. Vidal, T. Mano, N. Ha, T. Kuroda, M. V. Durnev, M. M. Glazov, E. L. Ivchenko, X. Marie, T. Amand, K. Sakoda, G. Wang, and B. Urbaszek, “Charge tuning in [111] grown GaAs droplet quantum dots”, Applied Physics Letters, vol. 105, no. 8, 082111/p.1-p.4, 2014
[6] X. Liu, N. Ha, H. Nakajima, T. Mano, T. Kuroda, B. Urbaszek, H. Kumano, I. Suemune, Y. Sakuma, and Kazuaki Sakoda, “Vanishing fine-structure splittings in telecommunication-wavelength quantum dots grown on (111)A surfaces by droplet epitaxy”, Physical Review B, vol. 90, no. 8, 081301(R)/p.1-p.6, 2014
[7] M. Bollani, S. Bietti, C Frigeri, D. Chrastina, K. Reyes, P. Smereka, J. M. Millunchick, G. M. Vanacore, M. Burghammer, A. Tagliaferri, S. Sanguinetti, “Ordered arrays of embedded Ga nanoparticles on patterned silicon substrates”, Nanotechnology, Vol. 25, No. 20, 205301, 2014
[8] S. Bietti, C. Somaschini, L. Esposito, A. Fedorov, S. Sanguinetti, “Gallium surface diffusion on GaAs (001) surfaces measured by crystallization dynamics of Ga droplets”, Journal of Applied Physics, Vol. 116, No. 11, 114311, 2014
[9] E. M. Sala, M. Bollani, S. Bietti, A. Fedorov, L. Esposito, S. Sanguinetti, “Ordered array of Ga droplets on GaAs(001) by local anodic oxidation”, Journal of Vacuum Science & Technology B, Vol. 32, No. 6, 061206, 2014
[10] Neul Ha, Takaaki Mano, Takashi Kuroda, Kazutaka Mitsuishi, Akihiro Ohtake, Andrea Castellano, Stefano Sanuinetti, Takeshi Noda, Yoshiki Sakuma, and Kazuaki Sakoda, “Droplet epitaxy growth of telecom InAs quantum dots on metamorphic InAlAs / GaAs(111)A”, Japanese. Journal of Applied Physics, vol. 54, 04DH07/p.1-p.3, 2015
[11] S. Bietti, J. Bocquel, S. Adorno, T. Mano, J. G. Keizer, P. M. Koenraad, S. Sanguinetti, “Precise shape engineering of epitaxial quantum dots by growth kinetics”, Physical Review B, Vol. 92, No. 7, 075425, 2015
[12] H. Kumano, H. Nakajima, T. Kuroda, T. Mano, K. Sakoda, and I. Suemune, “Nonlocal biphoton generation in a Werner state from a single semiconductor quantum dot”, Physical Review B, Vol. 91, No. 20, 205437/p.1-p.6, 2015
[13] Neul Ha, Takaaki Mano, Ying-Lin Chou, Yu-Nien Wu, Shun-Jen Cheng, Juanita Bocquel, Paul M. Koenraad, Akihiro Ohtake, Yoshiki Sakuma, Kazuaki Sakoda, and Takashi Kuroda, “Size-dependent line broadening in the emission spectra of single GaAs quantum dots: Impact of surface charges on spectral diffusion”, Phys. Rev. B, Vol. 92, No. 7, 075306/p.1-p.8, 2015
[14] Shi-qiang Li, Kazuaki Sakoda, John B. Ketterson, and Robert P. H. Chang, “Broadband

resonances in indium-tin-oxide nanorod arrays", Applied Physics Letters, Vol. 107, 031104/1-5 (2015).
[15] S. Bietti, L. Esposito, A. Fedorov, A. Ballabio, A. Martinelli, S. Sanguinetti, "Characterization and Effect of Thermal Annealing on InAs Quantum Dots Grown by Droplet Epitaxy on GaAs(111)A Substrates", Nanoscale Research Letters, Vol. 10, 247/p.1-p.7, 2015
[16] Takaaki Mano, Kazutaka Mitsuishi, Neul Ha, Akihiro Ohtake, Andrea Castellano, Stefano Sanguinetti, Takeshi Noda, Yoshiki Sakuma, Takashi Kuroda, and Kazuaki Sakoda, "Growth of Metamorphic InGaAs on GaAs (111)A: Counteracting Lattice Mismatch by Inserting a Thin InAs Interlayer", Crystal Growth & Design, Vol.16, No.9, 5412-5417 (2016).
[17] Hidekazu Kumano, Takumi Harada, Ikuo Suemune, Hideaki Nakajima, Takashi Kuroda, Takaaki Mano, Kazuaki Sakoda, Satoru Odashima, and Hirotaka Sasakura, "Stable and efficient collection of single photons emitted from a semiconductor quantum dot into a single-mode optical fiber", Applied Physics Express, Vol. 9, 032801/1-4 (2016).
[18] Neul Ha, Takaaki Mano, Yu-Nien Wu, Ya-Wen Ou, Shun-Jen Cheng, Yoshiki Sakuma, Kazuaki Sakoda, and Takashi Kuroda, "Wavelength extension beyond 1.5μm in symmetric InAs quantum dots grown on InP(111)A using droplet epitaxy", Applied Physics Express 9, 101201/1-4 (2016).
[19] M. Vidal, M. V. Durnev, L. Bouet, T. Amand, M. M. Glazov, E. L. Ivchenko, P. Zhou, G. Wang, T. Mano, T. Kuroda, X. Marie, K. Sakoda, and B. Urbaszek, "Hyperfine coupling of hole and nuclear spins in symmetric (111)-grown GaAs quantum dots", Phys. Rev B, Vol.94, 121302(R)/1-5 (2016).
[20] Patchareewan Prongjit, Somchai Ratanathammaphan, Neul Ha, Takaaki Mano, Kazuaki Sakoda, and Takashi Kuroda, "Type-II recombination dynamics of tensile-strained GaP quantum dots in GaAs grown by droplet epitaxy", Appl. Phys. Lett., Vol. 109, 171902 (2016).
[21] F. Biccari, S. Bietti, L. Cavigli, A. Vinattieri, R. Notzel, M. Gurioli, S. Sanguinetti, "Temperature activated coupling in topologically distinct semiconductor nanostructures", J. Appl. Phys. Vol. 120, No.13, 134312, 2016
[22] R. Bergamaschini, S. Bietti, A. Castellano, C. Frigeri, C. V. Falub, A. Scaccabarozzi, M. Bollani, H. von Kanel, L. Miglio, S. Sanguinetti, "Kinetic growth mode of epitaxial GaAs on Si(001) micro-pillars", J. Appl. Phys. Vol. 120, No. 24, 245702, 2016
[23] Natalia N. Schlenskaya, Yuanzhao Yao, Takaaki Mano, Takashi Kuroda, Alexey V. Garshev, Vadim F. Kozlovskii, Alexander M. Gaskov, Roman B. Vasiliev, and Kazuaki Sakoda, "Scroll-like alloyed CdSxSe1-x nanoplatelets: facile synthesis and detailed analysis of tunable optical properties", Chem. Mater., Vol. 29, 579-586 (2017).
[24] D. Scarpellini, A. Fedorov, C. Somaschini, C. Frigeri, M. Bollani, S. Bietti, R. Noetzel, S. Sanguinetti, "Ga crystallization dynamics during annealing of self-assisted GaAs nanowires", Nanotechnology, Vol. 28, No. 4, 045605, 2017
[25] Takashi Kuroda, "Entangled photon pair generation in naturally symmetric quantum dots grown by droplet epitaxy" in Micro- and Nanophotonic Technology edited by P. Meyrueis, K. Sakoda, M. Van de Voorde (Wiley, 2017) p.103-p.124

２．学会発表

[1] 【招待講演】Kazuaki Sakoda, "Photonics research activity at NIMS", Nano Korea 2015, COEX, Seoul, 2015/7/1
[2] 間野高明,「GaAs(111)A 基板上の高対称性量子ドット LED の作製」, GaAs(111)A 基板上の高対称性量子ドット LED の作製」, 名古屋国際会議場, 2015/09/13
[3] 【招待講演】間野高明,「(111)A 基板上の格子不整合系エピタキシーの諸現象：ドット形成と歪み緩和の理解と制御にむけて」, 第１１回量子ナノ材料セミナー , 電気通信大学（調布）, 2015/12/08
[4] Takashi Kuroda, 「Robustness characterization of entangled-photon emitters based on

naturally-symmetric droplet quantum dots」, The 9th Int. Conf. on Quantum Dots (QD2016), Jeju Ramada Plaza Hotel, Korea, 2016/5/22

[5] 黒田隆,「量子ドットもつれ光子源の安定性について 」, 第 77 回応用物理学会秋季学術講演会，朱鷺メッセ，新潟，2016/9/13

[6] 間野高明,「InGaAs/InAs/GaAs(111)A 成長時の格子緩和機構」, 第 77 回応用物理学会秋季学術講演会，朱鷺メッセ，新潟，2016/9/13

３．ワークショップ・セミナー・シンポジウム等の開催

[1] 2014 年 12 月 11 日，ミラノ・ビコッカ大学 物質科学科主催,「FemtoTera Kick Off Meeting」，ミラノ・ビコッカ大学（イタリア，コモ）

[2] 2015 年 11 月 5 日，物質・材料研究機構 先端フォトニクス材料ユニット主催,「第 106 回 APM セミナー」，物質・材料研究機構（つくば）

[3] 2016 年 9 月 29 日～2016 年 9 月 30 日，ハンガリー科学アカデミー 応用物理・物質科学研究所主催,「FemtoTera Workshop」，オーブダ大学（ハンガリー，ブダペスト）

４．研究交流の実績

[1] 2014 年 12 月 10 日～2014 年 12 月 14 日，迫田和彰（物質・材料研究機構），ミラノ・ビコッカ大学（イタリア，コモ），キックオフミーティング参加と研究所見学

[2] 2014 年 12 月 10 日～2014 年 12 月 14 日，黒田隆（物質・材料研究機構），ミラノ・ビコッカ大学（イタリア，コモ），キックオフミーティング参加と研究所見学

[3] 2015 年 10 月 31 日～2015 年 11 月 13 日，Akos NEMCSICS（オーブダ大学），物質・材料研究機構（つくば），研究打合せ

[4] 2015 年 11 月 2 日～2015 年 11 月 8 日，Stefano SANGUINETTI（ミラノ・ビコッカ大学），物質・材料研究機構（つくば），研究打合せ

[5] 2015 年 11 月 2 日～2015 年 11 月 8 日，David SCARPELLLINI（ミラノ・ビコッカ大学），物質・材料研究機構（つくば），研究打合せ

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[7] 2015 年 11 月 2 日～2015 年 11 月 8 日，Michele DEVETTA（ミラノ工科大学），物質・材料研究機構（つくば），研究打合せ

[8] 2016 年 9 月 26 日～2016 年 10 月 3 日，黒田隆（物質・材料研究機構），オーブダ大学（ハンガリー，ブダペスト），ワークショップ参加

[9] 2016 年 9 月 26 日～2016 年 10 月 3 日，間野高明（物質・材料研究機構），オーブダ大学（ハンガリー，ブダペスト），ワークショップ参加

５．特許出願

無し

６．受賞等

無し

７．その他

無し