Final report
of the **NN114457** (Concert-Japan FemtoTera ERA-NET) project

Project title: **Plasmon-enhanced Tera-Hertz emission by Femtosecond laser pulses of nanostructured semiconductor/metal surfaces**

Project duration: from **2014 November 01** to **2017 October 31**.

Description of the project aim

The aim of the project was the fundamental technological field of ultrashort pulsed THz radiation generation for time domain spectroscopy. This is an important reserch field, due to the specific properties of THz radiation and its various applications. One of the effective way of the THz generation is femtosecond near-infrared laser irradiation on nano-structured semiconductor/metal surfaces. The project investigated the THz emission process at nano-structured surface by stimulating with ultrashort laser pulses. Here, the nano-engineered surface system means self-assembled semiconductor quantum-dot-molecules with metal-nano-particles.

The surface is nanostructured with bottom-up self-assembling processes. The applied fabrication process was the so called droplet epitaxy. Droplet epitaxy technique offers unique opportunity to realize both quantum-dot-molecules and metal-nano-particles with the same technology and with the same platform.

Here, the interaction between ultrashort laser pulses and the nano-structured surface has been investigated, both theoretically and experimentally, too. In the frame of the project, the FemtoTera consortium (with Japan, Italian and Hungarian participants) has defined and characterized in detail the quantum-dot-molecule with metal-nano-particle system to meet THz generation requirements by means of experimental and theoretical approaches. The quantum confined state energy and its localization and spatial matching of the coupled nano-structure systems were studied. Accurate morphological and optical characterization has permitted to understand the coupled nano-structure system. Furthermore, the growth process of these nano-structures and their physical properties in view of their use for efficient THz generation were investigated.

Milestones of the project

Four main tasks are targeted during the project, addressing the development of a novel THz generation technology, which is listed below:

Milestone 1: Development of bottom-up nano-structured semiconductor surface with coupled quantum-dot-molecule and metal-nano-particle systems by an innovative fabrication schemes called droplet epitaxy. The technique is based on advanced molecular beam epitaxial procedure for the self assembly and self-alignment of quantum-dot-molecule and metal-nano-particle.

Milestone 2: The investigation of plasmon-enhanced emission from the intraband transitions that takes place, when irradiated with femtosecond laser pulses, on surfaces functionalized with self-assembled quantum-dot-molecule coupled to metal-nano-particle.

Milestone 3: The nano-engineering of the quantum-dot-molecule coupled with metal-nano-particle system to meet THz generation requirements, in terms of intensity and frequency.

Milestone 4: Investigation of THz generation from semiconductor surfaces functionalized with nano-structure complex under near-infra-red ultrafast laser irradiation

In addition this project has a clear networking purpose to allow for the constitution of an international team of scientific research groups, located in different countries (Japan, Italy, Hungary), working in different areas of photonics and nano-structures. The expected added value of such network is the related to the possibility, given by the diverse origin of the groups involved, to address the complexity inherent in the developing a completely novel technology

Characterization of the nano-structure complex

The detailed analysis of the observed dependencies of the quantum-dot-molecule 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 quantum-dot-molecule electronic states. The atomic force microscopy image of the best quantum-dot-molecule configuration is shown on Fig. 1.

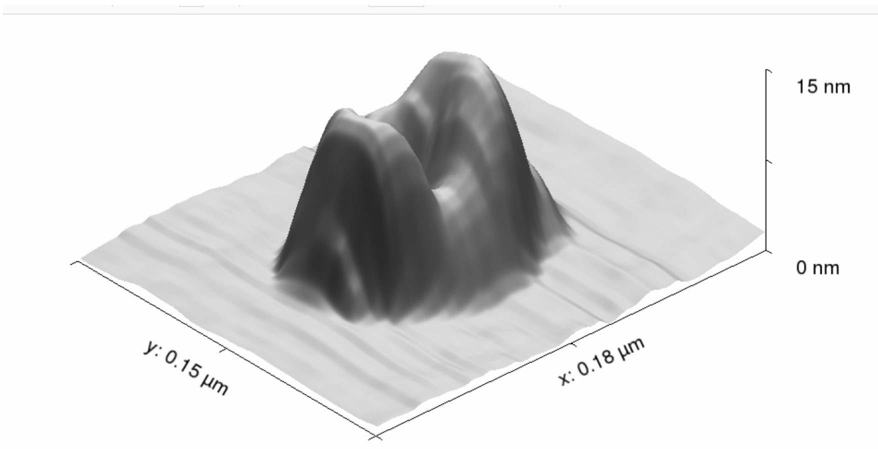

Fig. 1. *Atomic force microscopical image of a quantum-dot-molecule*

Transmission electron microscopy measurements demonstrate the achievement of QD complexes elongated in the [0-11] directions. For this measurement, heterogeneous crosssectional specimens must be thinned. Thin specimen can be achieved by mechanical and ion beam thinning methods, which is based mainly on the ion milling method. The sample preparation has crucial importance to achive clear transmission image. Thin cross-sectional specimens from the sample were prepared for the transmission electronmicroscopical investigations by cutting, embedding into a special holder, mechanical grinding, polishing, and finally ionbeam milling with 10 keV Ar ions. The conventional imaging of the samples was performed on a Philips CM20 electron-microscope at 200 kV. The high resolution images were taken with 300 kV JEOL 3010 high resolution microscope.

The conventional and the high-resolution transmission electronmicroscopical pictures of the nanostructures are shown in two directions which are perpendicular to each other. On the Fig. 2 and Fig. 3. are the nanostructures cutted in x and y directions, respectively. Where the A and B parts of each figures show the identical nanostructure in different magnificationtion.

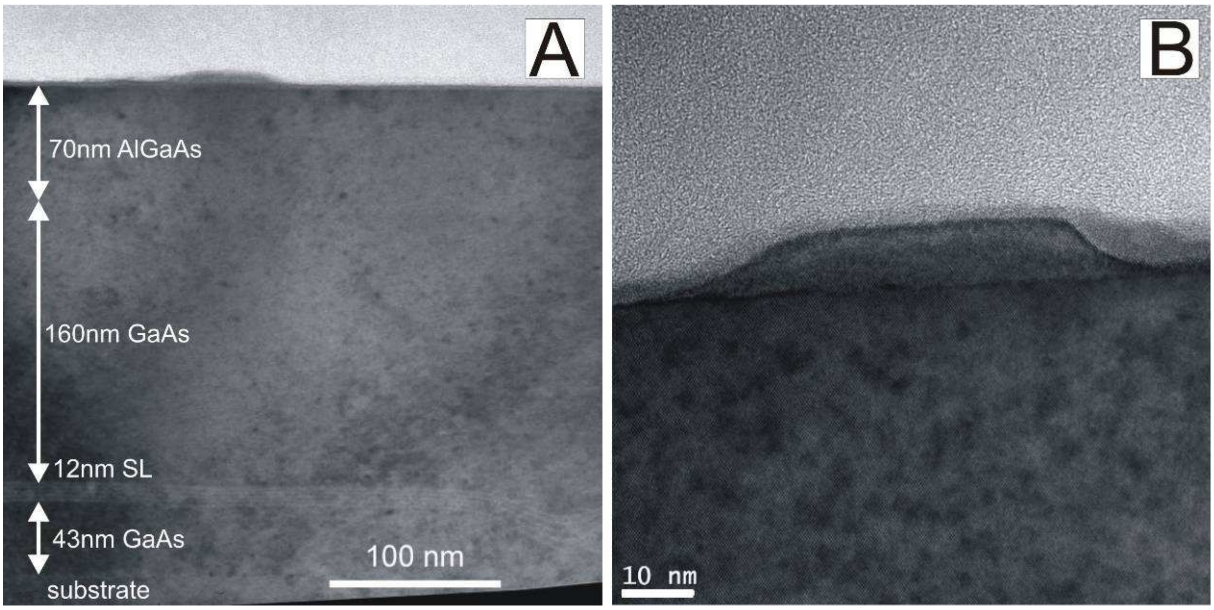

Fig. 2. *The A and B parts of the figure show conventional and high-resolution TEM image in x-direction, respectively. The pictures (A and B) show identical nanostructure in different resolutions.*

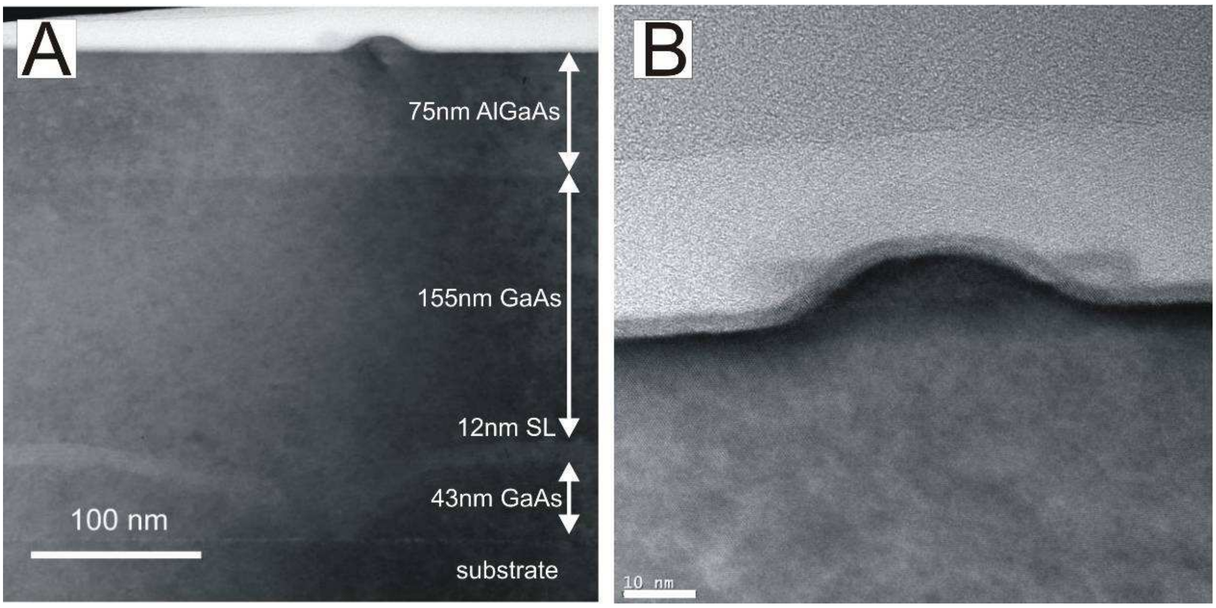

Fig. 3. *The A and B parts of the figure show conventional and high-resolution TEM image in y-direction, respectively. The pictures (A and B) show identical nanostructure in different resolutions.*

The part A and B of pictures show conventional and high-resolution image, respectively. The investigated cross-sections are perpendicular {110} directions on each other. The cross sectional high-resolution TEM image of the droplet epitaxial nano-structure shows that the nano-structure is perfectly crystalline. The crystal planes of the substrate are continued without any break in the nanostructure, no defects can be observed at the interface. The darker contrast on the structure edge corresponds with the nanometer sized GaAs capping layer.

The electrionic structure of the quantum-dot-molecule was modelled. The calculated electronic structure helped us to optimize the shape of the nano-structure. The electronic wave functions of the first two electron and hole states are shown on Fig. 4.

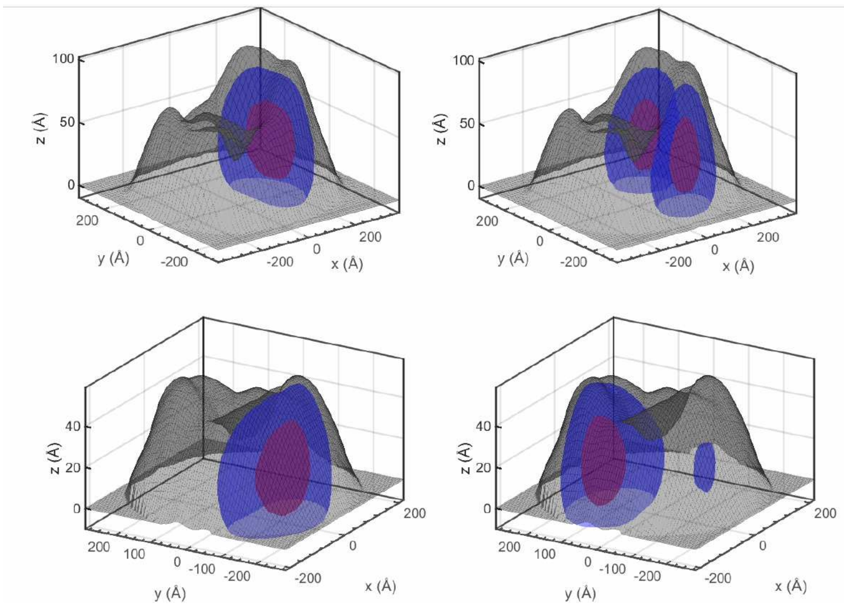

Fig. 4. *Electronic wave functions of the first two electron and hole states.*

The quantum-dot-molecules were investigated optically, too. Fig. 5 shows the micro photoluminescence spectra of a quantum-dot-molecule. Different spectra were confirmed depending on the monitoring position. Each spectrum consists of a few numbers of spectral lines that are close to each others, being associated with recombination from a single isolated quantum-dot-molecule. This observation therefore supports that each nano-structue has a fine energy structure with a typical splitting energy of several meV. The energy splitting agrees with the ground state - excited state splitting of the electron state calculated for the model quantum-dot-molecule structures.

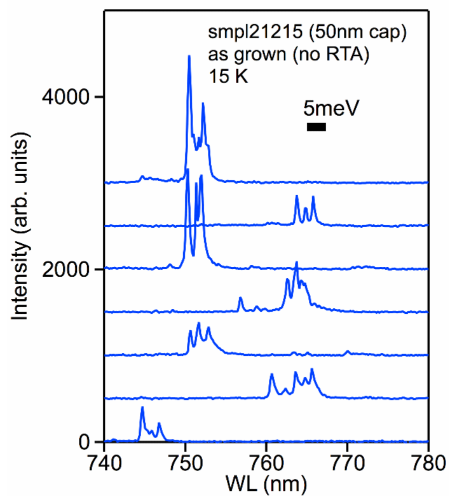

Fig. 5. *Micro photoluminescence spectra of the quantum-dot-molecule.*

To achieve appropriate formation of nano-structure complex of quantum-dot-molecule with metal-nano-particle, we used the same self-assembly procedure based on metallic droplets that was used for the quantum-dot-molecule fabrication, which was based also on droplet epitaxy. By removing the non-metallic group background pressure in the growth chamber it is possible to deposit gallium droplet with an independent control over density and size over a native surface. The role of the substrate temperature and gallium and arsenic fluxes, during the quantum-dot-molecule fabrication and the metallic-nano-particle alignment is fundamental. The relative positioning of these nano-structures can be only obtained through a careful control of gallium droplet nucleation sites via its surface diffusion and deposition flux (Fig. 6.). Fundamental, due to the nano-structure complex design imposed by the application, is for the metal-nano-particle to sit in between the two dots. This requires a fine tuning of the temperature. Also the gallium flux plays a role, allowing for the matching of the densities of the quantum-dot-molecules and metal-nano-particle.

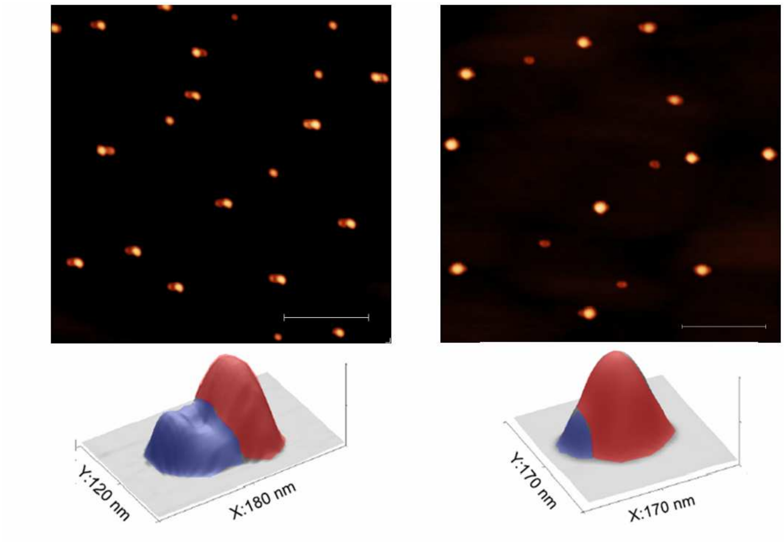

Fig. 6. *Positioning optimization of the gallium nano-particle (upper part: top view, lower part: single nano-structure complex in perspectivic view).*

Finally, the THz emission was investigated.. The measuring apparatus was driven by an amplified laser system that delivers 25 fs pulses with a bandwidth of 60 nm centered at 800 nm. The THz detection beams are obtained by splitting the incoming 1.6 W/800nm beam. Experiments carried out on the samples of nano-structure complexes, which show that the THz probe beam is substantially absorbed by the excited GaAs substrate. The transmission geometry thus emerges as not suitable for the measure of the THz field emitted by the gallium droplet on GaAs quantum molecules as it will be totally absorbed by the substrate during the experiment.

Project outcomes

The project fulfilled all the milestones planned. The targeted quantum-dot-molecule with metal-nano-particle complex configuration was obtained within the second round, thus making the planned third run not necessary and the reach of milestone 3 included in the milestone 2. For all the deliverables, internal reports have been shared among the partners and the results will be the subject of scientific publications on peer-reviewed international journals.

FemtoTera workshop in Budapest:

A sientific workshop was held in the frame of our project. The location was Óbuda University, Budapest, Hungary. The event was from September 29 to 30, 2016. The opening lecturer was Prof. Norbert Kroo, member of the Hungarian Academy of Sciences.

The invited speakers and their lectures were the following:

Christian Heyn, Dr. (University of Hamburg) : Droplet Etching of Nanoholes for Quantum Dots (QDs), QD-Molecules, and QD-Plasmonic Hybrids

David Jesson, Prof. (Cardiff Univerity): Controlling Surface Chemical Potential with Liquid Droplets

Takashi Kuroda, Dr. (NIMS, Tsukuba): Quantum entangled photon-pairs from semiconductor quantum dots

Takaaki Mano, Dr. (NIMS, Tsukuba): Lattice-mismatched epitaxy on (111)A substrates: Formation of quantum dots and metamorphic layers

Emmanouil Dimakis, Dr. (Helmholtz-Zentrum Dresden-Rossendorf): The role of Ga droplets in the epitaxy of GaAs nanowires on Si substrates

Sergio Bietti, PhD (L-NESS Laboratory, Como): Topic of the contribution was droplet epitaxy

Cristian Manzoni, PhD (CNR Dipartimento di Fisica - Politecnico di Milano): Topic of the contribution is laser spectroscopy

Stefano Sanguinetti, Ass. Prof. Dr. (Università degli Studi di Milano-Bicocca): Topic of the contribution is droplet epitaxy

Ákos Nemcsics, Prof. (University Óbuda, Inst. Micoel. and Technol. , Budapest): Capability of the droplet epitaxy in the view point of quantum circuits

Antal Ürmös, PhD Student (University Óbuda, Inst. Micoel. and Technol. , Budapest): Self organizing map for the technology support of droplet epitaxy

Fig. 7. *Part of the invited lecturers*

More informations are available on the home page: http://femtotera.uni-obuda.hu/

Related publications appeared during the project:

Ákos Nemcsics: Quantum Dots Prepared by Droplet Epitaxial Method; In: Quantum Dots - Theory and Applications; Ed.: Vasilios N Stavrou, Rijeka: InTech, Chapter 5; (2015) pp 119-149 (ISBN:978-953-51-2155-8)

Ákos Nemcsics: On the shape formation of the droplet epitaxial quantum dots; *Microelectronics Reliability* 56, pp. 73-77. (2016)

Ákos Nemcsics, Bálint Pődör, Lajos Tóth, János Balázs, László Dobos, János Makai, Márton Csutorás, Antal Ürmös: Investigation of MBE grown inverted GaAs quantum dots; *Microelectronics Reliability* 59, pp. 60-63. (2016)

Péter Kucsera, Tamás Sándor, Gusztáv V. Tényi, Márton Csutorás, Gergő Bátori, Béla Kupás-Deák, István Réti, Antal Ürmös, Ákos Nemcsics: Nanostructure Growth Supported by In Situ RHEED Evaluation; *Materials Science Forum* 885, pp. 234-238. (2017)

Ákos Nemcsics: Some aspects to the understanding of the droplet epitaxial nano-hole formation; *Journal of Crystal Growth* 477 pp. 2-6. (2017)

Antal Ürmös, Zoltán Farkas, Tamás Sándor, Ákos Nemcsics: Soft-computing based classification and design of quantum dot nanostructures on GaAs substrate; IEEE 15th International Symposium on Intelligent Systems and Informatics : SISY 2017. Szabadka, Szerbia, 2017.09.14-2017.09.16. New York: IEEE, 2017. pp. 123-127. (ISBN:978-1-5386-3855-2)

Antal Ürmös, Zoltán Farkas, Ákos Nemcsics: Modeling of III-V-based Nanohole Filling; *Acta Polytechnica Hungarica* **17** pp. 91-111. (2017)

Antal Ürmös, Zoltán Farkas, Ákos Nemcsics: Contribution to the understanding of III-V based nanohole filling; *American Journal of Condensed Matter Pphysics* **7** pp. 50-56. (2017)

Antal Ürmös, Zoltán Farkas, Márk Farkas, Tamás Sándor, László T Kóczy, Ákos Nemcsics: Fuzzy and Kohonen SOM based classification of different 0D nanostructures; SAMI 2017 : IEEE 15th International Symposium on Applied Machine Intelligence and Informatics. 510 p. Herlany, Szlovákia, 2017.01.26-2017.01.28. Budapest: IEEE, 2017. pp. 365-369. (ISBN:978-1-5090-5654-5)

Related publications under appearing:

Sergio Bietti, Francesco B. Basset, David Scarpellini, Alexey Fedorov, Andrea Ballabio, Luca Esposito, Martin Elborg, Takashi Kuroda, Akos Nemcsics, Cristian Manzoni, Caterina Vozzi, Stefano Sanguinetti: Nano-positioning of Ga droplets on GaAs Quantum Molecule nanostructures (manuscript is ready, intended journal: Applied Physics Letters)

Ákos Nemcsics: Droplet epitaxy as a tool for the QD based circuit realization; : InTech chapter of Quantum Dot (accepted, under print)

Antal Ürmös, Zoltán Farkas, Márk Farkas, Tamás Sándor, László T. Kóczy, Ákos Nemcsics: Application of Self-Organizing Mapping for Technological Support of Droplet Epitaxy; *Acta Polytechnica Hungarica* (accepted, under print)

Antal Ürmös, Zoltán Farkas, Lajos Tóth, Ákos Nemcsics: Investigation of Droplet Epitaxially Grown Inverted Quantum Dot; presented on the conference of OATK2017 (XI. Országos Anyagtudományi Konferencia) 15-17 Oktober 2017, Balatonkenese (appearance in abstract booklet, manuscript under preparation)

Ákos Nemcsics, Lajos Tóth, Zoltán Erdélyi: Particular behaviour of the GaAs wetting layer on AlGaAs substrate during dropet epitaxy; presented on the conference of ECOSS33 (33rd European Conference on Surface Science) 27 August – 01 September 2017 Szeged (appearance in abstract booklet, manuscript under preparation)