Publishable summary:

The principal aim of this Marie-Curie European Reintegration Grant 'SUPERRAD' no 271640 was to foster a long-term reintegration of the fellow - Jacek Kasprzak - within the host institution (HI) - Institut Néel CNRS Grenoble. This goal was attained far beyond expectations. The fellow joined HI in 2010 as a permanent research scientist. Having in mind the know-how he gained upon his previous Marie-Curie training (Marie-Curie IEF CUSMEQ no 217962, 2008-2010) he was expected to develop previously non-existing research field at HI: coherent nonlinear spectroscopy of semiconductor nanostructures. The first funding obtained within this ERG grant allowed him to make an initial first step to set up the scientific laboratory of coherent nonlinear optical spectroscopy. His activity attracted in 2012 a master student (currently a PhD student in the group of the Fellow), who prepared a diploma thesis regarding active pulseshaping of short laser pulses employing spatial light modulators. In 2012 the fellow was awarded ERC Starting Grant 'PICSEN', which dramatically boosted the scientific development of the Fellow: allowed to fully equip the lab in 2013 and achieve in 2014 the world-class results in the field of coherent spectroscopy of individual emitters in solids.

The developed spectrosopic technique is based on heterodyne spectral interferometry (Langbein & Patton, Optics Letters 2006). In this approach, one uses short (sub-ps) optical pulses: E1, E2 and E3, so as to resonantly drive a nonlinear response of an exciton (a dipole moment created by an electron-hole pair, here trapped in a semiconductor quantum dot), like four-wave mixing (FWM), which is proportional to E1*E2E3. Measuring FWM as a function of time delay (within a ns range) between the pulses E1 and E2 yields exciton's coherent dynamics, while varying the E2-E3 time delay yields population lifetime. This experimental tool enabled within last years pioneering investigations of cavity quantum electrodynamics effects in solids (Kasprzak Nature Materials 2010, Albert Nature Communications 2013) and inter-exciton coherent couplings (Kasprzak Nature Photonics 2010). The new setup outperforms the previous generation of this technique - until recently exclusively available in the group of Prof. W. Langbein at Cardiff University UK - in several aspects. Some of important ones are: i) a versatile heterodyne mixing, which allows to access various types of optical nonlinearities (non-degenerate FWM, six-wave mixing, etc.), ii) introducing of E2-E3 delay enabling to measure in non-degenerate configurations, iii) control over the spatial position between E1 and E3 beams along with wide range of spectral and spatial imaging, which paves the way towards investigations of spatial propagation of coherence. Setting up this technique at HI opens many exciting prospects for experiments within a wide range of new materials and places the Fellow in years to come as an important player in the field of coherence in condensed matter.

Scientific objectives of the project

The direct scientific objective of the project was demonstration of the superradiance (SR) effect - self-organized build up of coherent radiation within an ensemble of quasi-degenerate emitters of light - in a semiconductor nanostructure.

Work progress

The above scientific objective has not been achieved within the project's timescale and time commitment of the Fellow (25% over 3 years). The proposed calendar turned out not to be fully realizable due to a two-fold reason. First, the funds provided by the ERG project only partially could have covered equipment costs necessary to assemble the experimental setup. Second, the first samples: GaAs layers of high purity containing donor-bound excitons, required for the realization of the project, have become available only in the third year of the project in the framework of new collaboration with Toulouse (Ch. Fontaine and A. Arnault, Laboratoire d'Analyse et d'Architecture des Systèmes). Furthermore, in the third year of the project the Fellow's activity strongly deviated towards realization of the ERC Starting Grant 'PICSEN' (launched in December 2012). The equipment listed in the proposal (B3.3) was purchased (apart from the lock-in amplifier, bought instead in the framework of PICSEN and HeNe laser, which turned out not to be indispensable). Conversely, more lab consumables, primarily opto-mechanics, and small lab equipment (laser power meter, infrared viewer) have been purchased.

Research progress:
We would like to point successful accomplishment of several intermediate tasks proposed in the description of work, which are particularly relevant in the further research for the SR effect in semiconductor nanostructures:

1. ***Spatio-temporal pulseshaping (Workpackage 2, Task 3)***

   A pulseshaper is an optical apparatus employed to engineer temporal transients of short (femto-second) optical pulses, so as to attain desirable duration, temporal chirp (temporal distribution of different spectral components) of the pulse and its spectral content.

   a. Construction of a passive pulseshaper.
      The schema and our realization of the passive pulseshaper are shown in Fig. 1a. A short laser pulse is dispersed by a diffraction grating, so that its frequency spectrum is imaged in the vicinity of the spherical mirror. The spectrum of the recompressed pulse can be shaped by varying the position of the apodizing slit (Fig. 1b) in front of the mirror, while the chirp (temporal sequence of spectral components within the pulse) can be tuned by varying the distance between the grating, the lens and the spherical mirror. In our practical realization (Fig. 1c), a Fourier limited 150fs pulse provided by a Ti:Sapphire laser Tsunami from Spectra Physics can be shaped (and chirped or pre-shirped) to attain a few ps. Such pulse-shaping and chirping with a passive pulseshaper is a necessary ingredient in coherent nonlinear spectroscopy, specifically in four-wave mixing of individual quantum dots currently developed by the Fellow at the HI.

   b. Construction of an active pulseshaper based on spatial-light modulator.
      The schema of the active pulseshaper is shown in Fig. 2a. Here, we performed spatio-temporal pulse shaping employing an active liquid crystal element. Specifically, we used a high resolution, two-dimensional spatial light modulator (SLM), model PLUTO-NIR provided by Holoeye. Our set-up consists of zero dispersion line with the SLM in a Fourier plane, so as to modify the phase of each spectral component composing the femtosecond pulse reflected from the SLM. We characterised our device and we have retrieved the phase shift of the reflected light as a function of driving voltage

applied to each pixel, as shown in Fig. 2b. Next, we have observed that an intrinsic spatio-temporal coupling of such a line, allow us to measure spatial modulation of the spectral components in real space by using periodical modulation of the shifted phase in the Fourier plane, as presented in Fig. 2c. As a next step, we will measure shaped pulses using an autocorrelator enhanced with a frequency-resolved optical gating (FROG), so as to retrieve temporal pulse shape. Future developments, directly linked with the ERC StG PISCEN project, will consist in applying shaped pulses for coherent spectroscopy of individual emitters in semiconductor nanostructures. In particular, such a pulse shaping in the frequency domain is required for coherent control of excitons in quantum dots, commonly characterized by a sub-nanosecond dephasing.

2. ***Realization of the micro-photoluminescence hyperspectral imaging setup (Workpackage 1, Task 2).***

Hyperspectral imaging techniques yield spectrally resolved linear (fluorescence, photoluminescence, reflectivity, absorption) or non-linear (four-wave mixing, CARS) optical response of the investigated sample as a function of the spatial position in one, two or three dimensions. An access to such a tool is a prerequisite for demonstration of the SR effect in the nanostructures, as it enables to assess spectral and spatial density and distribution of individual emitters. This crucial tool could only have been constructed by the Fellow at the end of the SUPERRAD project, having at hand financial means of the ERC PICSEN project. The core of the setup is shown in Fig. 3a. It consists of the cryostat, which allows performing experiments in the range 5K-300K. Due to a relatively small binding energy of individual light emitters considered here (donor-bound excitons, excitons in semiconductor quantum dots) are optically active at low temperatures. For this reason our experiments are carried out at 5K. The excitation laser is focussed via the high NA objective at the sample surface reaching diffraction limit. The optical response is spectrally resolved via imaging spectrometer (ACTON 2750i) and recorded on the chip of the CCD camera (PIXIS 400 eXcelon). In order to create hyperspectral images, we scan the sample position (up to 10x10mm with the resolution of 50nm), employing close-loop encoded stepper motors provided by PIMicos, which are installed inside the cryostat. Conversely, we can also move the position of the objective (up to 0.25x0.25x0.4mm, 1nm resolution), which is set on the three-dimensional close-loop piezo stage provided by Physik Instrumente. An example of the hyperspectral images obtained on CdTe/ZnTe quantum dots (provided by collaborators from Warsaw University, growth by W. Pacuski) is shown in Fig. 3b. The emission from individual quantum dots for a given energy can be observed. In Fig. 3c we present hyperspectral imaging of ultra-low density GaAs/AlGaAs quantum dots obtained by unintentional formation of gallium droplets (growth by R. Planel, Paris). We observe the emission from only several dots within the spatial range of 135x135 microns. Additionally, the scattered light reproduces the topographic state of the sample surface. In Fig. 4a we present hyperspectral image of high purity GaAs layers, provided by A. Arnault from LAAS Toulouse (Workpackage 1, Task 1), for the emission wavelength of donor-bound excitons 812.7nm. A pair of dark regions corresponds to dust on the sample surface appeared after cleaving of the wafer. The emission is rather homogenous and emission of individual states cannot be observed, indicating high density of donor-bound excitons. The averaged micro-photoluminescence spectrum reveals rich features. It is dominated by the donor-bound exciton transitions (D°X), but also emission from free excitons (FX), acceptor-bound excitons (A°X) and two-electron satellites (TES) can be identified (Workpackage 1, Task 2).

3. ***Realization of the spectral interferometry setup (Workpackage 2, Task 4).*** Spectral interferometry is the technique of nonlinear spectroscopy permitting the retrieval of amplitude and phase of the coherent optical response, using prior knowledge regarding arrival sequence of the signal and the reference field, which spectrally interferes with the signal. The spectral interferometry method combined with optical heterodyning paved the way towards measurement of coherent nonlinear responses of individual emitters in solids, like excitons in semiconductor quantum dots. In the SUPERRAD project we proposed to use spectral interferometry to observe temporal dynamics of the up-converted SR burst. The spectral interferometry was set up by the Fellow at the HI and is now used in single quantum dot wave-mixing experiments. An example of spectral interferogram of the four-wave mixing signal from individual InAs strongly-confined quantum dots is shown in Fig. 5.

Advancement beyond the state of the art:

The major advancements beyond the state of the art of have been performed in the field of coherent nonlinear spectroscopy. The Fellow has shown that photonic enhancement in weakly-coupled microcavity dramatically facilitate (by a factor of 10-20) the retrieval efficiency of coherent responses of single quantum dots. He performed single exciton four-wave mixing and six-wave mixing experiments on strongly-confined InAs quantum dots. He introduced non-degenerate configuration (three beams) in wave-mixing experiments. As concerns SR effect, no major advancements have been achieved as the Fellows activity strongly deviated towards coherent nonlinear spectroscopy.

Social impact

The realization of the SUPERRAD project highly contributed towards full reintegration of the Fellow at HI. The initial funding provided by the ERG allowed him to gained autonomy at HI and pursue his activity along his own original research axis and not to be incorporated instead into previously existing subjects in his new scientific environment. The grant allowed the Fellow to maintain very fruitful collaboration with Cardiff University and Warsaw University. It also triggered new collaborations (see Management section).

Scientific impact

The fellow demonstrated that single quantum dot wave-mixing experiments can be conducted outside the group of his previous post-doctoral adviser (prof. W. Langbein Cardiff University, UK), demonstrating broader feasibility of this steep and complex, yet powerful approach in coherent nonlinear spectroscopy. He demonstrated that by tailoring photonic environment of the semiconductor nanostructures the coherent nonlinear response of individual quantum systems can be retrieved more easily.

Transfer of knowledge

The Fellow fully transferred his skills and knowledge towards the HI. The heterodyne spectral interferometry technique permitting single exciton four-wave mixing experiments, previously available exclusively in the group of Prof. W. Langbein (Cardiff University UK), are now available at HI. Furthermore, in 2013-2014 the Fellow trained a post-doctoral researcher (F.

Fras), who has just obtained a permanent position (Maître de Conference) at the University of Strasbourg, and who will spread this knowledge further to his new host institution. The Fellow also contributed towards transfer of knowledge via presentations at conferences and seminars (see dissemination) and by training of the Master and PhD student (Q. Mermillod).

Additional information
As concerns demonstration of the SR effect in semiconductors, an important breakthrough has been achieved in 2012 in the group of J. Kono (Rice University, Houston, Texas, USA). In the paper published by G. T. Noe II et al. in Nature Physics **8**, p. 219 the authors report on defining features of the SR effect in dense electron-hole plasma in a quantum well under high magnetic field. By performing pump-probe measurements they observe a sudden drop of population and consecutive spontaneous emission in arbitrary direction of giant bursts of SR pulses. Importantly: i) the SR pulse is delayed with respect to the pump laser ii) the SR intensity scales with the square of number of self-organized dipoles contributing to formation of the SR. Further research for the SR of quantum dots aims for demonstrating this effect in semiconductor nanostructures in absence of external magnetic field.

Dissemination:

Publications:

J. Kasprzak, S. Portolan, A. Rastelli, L. Wang, J. D. Plumhof, O. G. Schmidt, and W. Langbein
Vectorial nonlinear coherent response of a strongly-confined exciton-biexciton system
New Journal of Physics 15, 055006 (2013).

Conferences 2011-2014:

8th International Conference on Quantum Dots, Pisa 2014, Italy
Q. Mermillod, F. Fras, G. Nogues, Ch. Hoarau, C. Schneider, M. Kamp, W. Langbein and J. Kasprzak
Cavity-enhanced coherent nonlinear response of a single strongly-confined InAs quantum dot
poster contribution

18th International Winterschool on New Developments in Solid State Physics, Mauterndorf 2014, Austria
session chair

French German Korean Workshop on Optics of Semiconductor Nanostructures 2013, Chamonix, France
F. Albert, K. Sivalertporn, J. Kasprzak, K. Sivalertporn, F.lbert, C. Schneider, S. Hofling, M. Kamp, A. Forchel, S. Reitzenstein, E. A. Muljarov and W. Langbein
On the quantum light-matter coupling in a semiconductor nanostructure
contributed talk

Journes Boîtes Quantiques 2013, Paris, France
F. Albert, K. Sivalertporn, J. Kasprzak, K. Sivalertporn, F.lbert, C. Schneider, S. Hofling, M. Kamp, A. Forchel, S. Reitzenstein, E. A. Muljarov and W. Langbein
On the quantum light-matter coupling in a semiconductor nanostructure
contributed talk

3rd International workshop on the optical properties of nanostructures, Bayreuth 2013, Germany
J. Kasprzak
Exploring coherence of individual emitters in a solid
invited talk

2nd workshop du GdR- IQFA, Grenoble 2012, France
F. Albert, K. Sivalertporn, J. Kasprzak, M. Straus, C. Schneider, S. Hofling, M. Kamp, A. Forchel,
S. Reitzenstein, E. A. Muljarov and W. Langbein
Microcavity controlled coherent coupling of quantum dots
contributed talk

12th International Conference on Physics of Light-Matter Coupling in Nanostructures, Hangzhou 2012, China
J. Kasprzak
Exploring coherence of individual emitters in a solid
invited talk

F. Albert, K. Sivalertporn, J. Kasprzak, M Strauss, C. Schneider, S. Hofling, M. Kamp, A. Forchel, S. Reitzenstein, E. A. Muljarov, and W. Langbein
Cavity-controlled coherent coupling in a cluster of excitonic qubits
poster contribution

41st Jaszowiec International School and Conference on the Physics of Semiconductors, Krynica 2012, Poland
J. Kasprzak
Exploring coherence of individual emitters in a solid
invited talk

F. Albert, K. Sivalertporn, J. Kasprzak, M Strauss, C. Schneider, S. Hofling, M. Kamp, A. Forchel, S. Reitzenstein, E. A. Muljarov, and W. Langbein
Cavity-controlled coherent coupling in a cluster of excitonic qubits

12th Conference on the Optics of Excitons in Confined Systems, Paris 2011, France
J. Kasprzak, B. Patton, V. Savona, W. Langbein
Coherent coupling between distant excitons revealed by two-dimensional nolinear hyperspectral imaging
oral contribution

E. A. Muljarov, K. Sivalertporn, J. Kasprzak and W. Langbein
Theory of the nonlinear optical response of a strongly coupled quantum dot-microcavity system
poster contribution

J. Kasprzak, L. Wang, J. D. Plumhof, S. Portolan, A. Rastelli, O. G. Schmidt and W. Langbein
Exploring the vectorial nonlinear coherent response of a strongly confined exciton in an individual quantum dot
poster contribution

J. Kasprzak, F. Albert, S. Reitzenstein, E. A. Muljarov, C. Kistner, C. Schneider, M. Strauss, S. Hofling, A. Forchel and W. Langbein
Climbing the polariton ladder of an exciton-cavity system
poster contribution

International Conference on Physics of Light-Matter Coupling in Nanostructures, Berlin 2011, Germany
J. Kasprzak, F. Albert, S. Reitzenstein, E. Muljarov, C. Kistner, C. Schneider, M. Strauss, S. Hofling, A. Forchel and W. Langbein
Climbing the polariton ladder of an exciton-cavity system
invited talk

Euro MBE, Alpe d'Huez 2011, France
J. Kasprzak, J.-S. Hwang, F. Donatini, C. Bougerol, H. Mariette, Le Si Dang and R. Songmuang
Correlating electronic and structural properties of Ga-assisted GaAs nanowires via cathodoluminescence imaging
poster contribution

Invited Seminars 2011-2014:
Coherent spectroscopy of nanostructures
October 2013, Warsaw University, Poland
invited by J. Szczytko in the framework of the "School on engineering of nanostructures".

On the quantum light-matter coupling in a semiconductor nanostructure
May 2013, IPCMS Strassburg, France invited by Prof. P. Gilliot

On the quantum light-matter coupling in a semiconductor nanostructure
March 2013, Warsaw University, Poland, invited by Prof. Jan Derezinski

On the quantum light-matter coupling in a semiconductor nanostructure
March 2013, Warsaw University, Poland, invited by Prof. Tadeusz Stacewicz

Exploring coherence of individual emitters in a solid
September 2012, Technishe Universitat, Wurzburg, Germany, invited by Dr. Sven Hofling

Exploring coherence of individual emitters in a solid
September 2012, Laboratoire Pierre Aigrain, Paris, France, invited by Prof. Carole Diederichs

Exploring coherence of individual emitters in a solid
July 2012, EPFL, Lausanne, Switzerland, invited by Prof. Benoit Deveaud

Coherent nonlinear spectroscopy under (high) magnetic field ?
May 2011, LNCMI, Grenoble, invited by Prof. Marek Potemski

Exploring coherence of individual emitters in a solid
April 2011, University of Warsaw, Poland, invited by Prof. Marian Grynberg

Outreach:

The Fellow regularly participates at the events promoting ERC and Marie-Curie Grants in Grenoble area, organized by GIANT, CNRS and CEA. Typical events are: preparatory meetings for ERC candidates, Action “Fostering ERC in Grenoble” and “Mock Interviews” for prospective ERC candidates.

Project Website: The HI created the website related to the project: http://neel.cnrs.fr/spip.php?article3225

Project Management:

The Fellow is fully reintegrated at HI, at the end of the fellowship he was promoted from the 2nd class to the 1st class research scientist at CNRS (stable permanent position). He is building his research group, which now is composed of 1 PhD and 1 post-doctoral researcher. He created his own research lab devoted to research in frontier issues of optical coherence in nanostructured semiconductors and condensed matter. He now holds the ERC Starting Grant entitled “Propagative and Internal Coherence in Semiconductor Nanostructures” (2012-2017, 1.5M€).

The fellowship was terminated according to the contract on 28/02/2014. From the management point of view the initial workplan was not completed and had to be revisited during the fellowship. The progress within the tasks was delayed due to the development of the new lab facilities (2011-2013). No gender or ethical issues to report.

Current active scientific collaborations:

Within the host institution: electronic engineer Ch. Hoarau, research scientist G. Nogues (hardware programming), members of NPSC team at Néel Institut.

Outside of the Host Instituton: CEA Grenoble, LAAS Toulouse, University of Milano, University of Warsaw, University of Cardiff, University of Wurzburg, Tyndall Institute (Cork, Ireland).