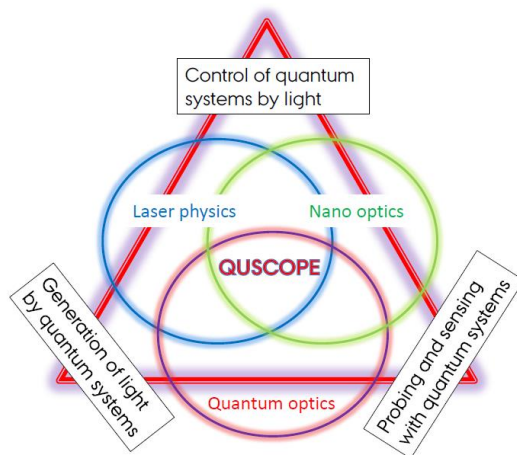


Villum Foundation Centre of Excellence

QUSCOPE



Midterm Report, December 2016

Introduction

The Villum Foundation Centre of Excellence, QUSCOPE, is a collaborative effort among the theoretical quantum optics, laser physics and nanooptics research groups at the University of Aarhus and the University of Aalborg in Denmark. The aim of QUSCOPE is to develop new proposals and solutions to the main challenges of the three respective research fields. This is accomplished with a focus on three overarching themes: *quantum dynamics induced and controlled by light*, *generation of light by quantum systems* and *probing of quantum dynamics by light* in physical systems ranging from atoms and molecules to solid state devices.

The Centre is in full operation, and with our remaining hiring plans we are in good agreement with our proposed 5-year budget. We shall submit the accounts for our spending in 2014-2016 as soon as the University Administrations have finished their accounting processes in January 2017.

On the following pages we present a summary of our achievements in the past three years and some of our plans for the following two years.

We wish to draw attention to the QUSCOPE webpage which is continuously updated with news, activities in the Centre, publications, staff and students, contact information, etc.:

<http://phys.au.dk/forskning/forskningsomraader/quscope/>

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Staff and students

QUSCOPE consists of the three group leaders, Klaus Mølmer, Thomas Garm Pedersen and Lars Bojer Madsen, and their postdocs and PhD students. The group leaders also host international internship students and they supervise bachelor and master thesis projects within topics related to the QUSCOPE research. The Centre disposes of part time assistance from a secretary and an IT-expert.

The QUSCOPE research teams receive funding from different additional sources, including the Universities of Aarhus and Aalborg, Danish national sources, EU and the US. We want all students and postdocs to benefit from (and contribute to) the QUSCOPE activities and projects, and we count all students and postdocs as members of QUSCOPE, irrespective of the funding sources covering their salaries and other expenses.

In the following we list all postdocs and PhD students employed in the research groups – (Q denotes if a person has been paid by the QUSCOPE grant).

Postdocs:

Malte C. Tichy, AU	October 1, 2011 – September 30, 2015
Haruhide Miyagi, AU	December 1, 2011 – November 30, 2016
Durga B. Dasari, AU	December 1, 2012 – January 31, 2016
Siddharta Chattopadhyay, AU	May 1, 2014 – April 30, 2016
Juan Omiste (Q), AU	August 1, 2014 – July 31, 2015 and February 1, 2016 – January 31, 2017
Qing Xu, AU	August 2014 – October 11, 2015
Camille Lévêque (Q – 2 years), AU	November 1, 2014 – October 31, 2017
Mads L. Trolle (Q), AAU	January 2015 – September 2015 and Oct 2016 – Dec 2016
Ralf Blattmann (Q), AU	April 1, 2015 – March 31, 2017
Stephen R. Power, AAU	Sep. 2015 – Mar. 2016
Daniel Reich, AU	September 1, 2015 – August 31, 2016
Tarek Elsayed (Q), AU	September 1, 2015 – August 31, 2017
Darko Dimitrovski (Q), AAU	September 2015 – September 2017
Yuan Zhang (Q), AU	September 1, 2015 – August 31, 2018
Felix Motzoi, AU	April 1, 2016 – March 31, 2018
Lukas Buchmann, AU	October 1, 2015 – September 30, 2017
Andrew Wade, AU	November 1, 2015 – April 30, 2016
Hector C. Mera, AAU	Aug. 2016 – Mar. 2017
Christian Kraglund Andersen, AU	August 1, 2016 – September 30, 2016

PhD students:

Christian Kraglund Andersen, AU	August 1, 2011– July 31, 2016
Andrew Wade, AU	May 15, 2012 – October 15, 2015 finished 16/4 2015
Yao-Chung Tsao, AAU	August 1, 2012 – July 31, 2016
Jens Bækthøj, AU	August 1, 2012 – July 31, 2016
Jens Svensmark, AU	February 1, 2013 – January 1, 2016
Lun Yue, AU	September 2013 – August 2016
Søren Bruun, AAU	September 2013 – August 2016
Morten R. Thomsen, AAU	September 2013 – August 2016

Kenneth Hansen (Q), AU	October 1, 2014 – September 30, 2018
Chuan Yu, AU	October 1, 2014 – September 30, 2017
Alexander Holm Kiilerich (Q), AU	August 1, 2014 – July 31, 2018
Eliska Greplova (Q), AU	November 1, 2014 – October 31, 2017
René Petersen, AAU	September 2014 – August 2017
Farzad Bonabi (Q), AAU	January 2015 – January 2018
Jinglei Zhang (Q), AU	April 15, 2015 – April 14, 2018
Qingli Jing (Q), AU	September 1, 2015 – July 31, 2018
Jørgen Johansen Rørstad (Q), AU	December 1, 2015 – November 30, 2018
Jonas Have (Q), AAU	Aug. 2016 – Aug. 2019
Philip Blocher, AU	August 2016 – July 2020
NN (Q), AAU	January 2017 – December 2020

Internships (visiting PhD students)

Marco Mattioli (Innsbruck, Austria)	August- December 2015
Peng Xu (Nanjing, China)	August 2016 – July 2017
Tahereh Abad (Tehran, Iran)	November 2016 – April 2017

QUSCOPE Centre Events

QUSCOPE has hosted a number of two-day retreats and one-day site visits in Aarhus and Aalborg for all members of the center. At these retreats and meetings, tutorials and exercise material serve to broaden the methods and ideas developed in the different research teams to the whole Centre. Courses have been offered in both Aalborg and Aarhus around the research themes of QUSCOPE. Visiting colleagues from abroad, e.g., for joint collaboration and as opponents for thesis exams in QUSCOPE, have offered seminars and lectures for QUSCOPE members and students. These seminars have also been well visited by other students in Aarhus and Aalborg. An updated list of events with abstracts and programs is provided on the QUSCOPE center homepage.

In Aarhus, QUSCOPE director, Klaus Mølmer, has organized meetings with colleagues from the departments of Engineering, Nanoscience, Computer Science and Mathematics, to strengthen the connections between research on quantum information and quantum technology in the departments and to coordinate input to the national FORSK2025 and the European Quantum Flagship initiatives. Joint scientific meetings with the new QMATH Villum Centre of Excellence in Copenhagen are currently being planned.

QUSCOPE theses

PhD:

Maciek Spiewanowski, *Orbital distortion and nondipole effects in strong field physics*, March 2014
 Yao-Chung Tsao, *Fabrication and Characterization of Thin-Film Amorphous Silicon on Metal Nanostructures*, September 2015
 Andrew Wade, *Manipulating collective quantum states of ultracold atoms by probing*, October 2015
 Lun Yue, *Strong-field-induced breakup of diatomic molecules*, March 2016
 Søren J. Brun, *Exploring the electronic and optical properties of semiconducting graphene*, September 2016

Morten R. Thomsen, *Electronic and magnetic properties of nanostructured graphene*, September 2016
Jens Egebjerg Bækthøj, *Attosecond Transient Absorption Spectroscopy of Atoms and Diatomic Molecules*, September 2016
Jens Svensmark, *Tunneling Ionization of Diatomic Molecules*, September 2016
Christian Kraglund Andersen, *Theory and design of quantum devices in circuit QED*, October 2016

MSc:

Jens Egebjerg Bækthøj, *Electron Dynamics: A Novel Numerical Approach and Attosecond Transient Absorption Spectroscopy*, May 2014
Jens Svensmark, *Strong Field Ionization of Small Molecules*, May 2014
Anders Larsen and Rolf Sommer Poulsen, *Applied Hartree-Fock methods*, June 2015
Chuan Yu, *Strong-Field Ionization of Helium*, December 2015
Joana Sohnesen, *The Stark effect in hydrogen* January 2016
Eliska Greplova, *Past Quantum States: Theory and Applications*, April 2016
Alexander Holm Killerich, *High sensitivity quantum measurements*, June 2016
Kenneth K. Hansen, *Two-Electron Correlation Effects in High Harmonic Generation*, September 2016
Jinglei Zhang, *Past Quantum State Theory for Gaussian States*, October 2016
Nikolaj Ravn, *Ionisationsdynamik induceret af infrarøde laserpulser*, will finish June 2017
Mathias Klahn, *To-step semiklassisk model for dynamik i stærke laser felter*, will finish June 2017

BSc:

Lars Mølgård Sørensen, *Om Anvendelse af Göppert-Mayer transformationen i forbindelse med stærkfeltsapproximationen*, 2014
Christian Kirk, *Using ion Coulomb crystals to generate exotic quantum effects*, 2014
Maja Juhl Lassen, *Kvantegates og kvantecomputing med fangede ioner*, 2015
Kristian Knakkegaard Nielsen, *Om målingsbetinget estimation af et kvantesystems baggrundsparemetre*, 2015
Philip Daniel Blocher, *Conditioned atom dynamics in optical detection*, 2015
Henrik Lund Mortensen, *Anharmonicity in superconducting resonators with a non-linear circuit embedded*, 2015
Nikolaj Ravn, *Dirac-ligningen og anvendelser for centralsymmetriske potentialer af ydre elektromagnetiske felter*, 2015
Emil Poulsen, *Simulating laser-induced breakup processes. A study of isotope effects in ionization and dissociation of H_2^+ and D_2^+ with intense laser pulses*, 2016
Magnus H. Geismar, *Relativistic Classical Monte Carlo Simulations of hydrogenlike ions in intense laser pulses*, 2016
Esben Mahler, *A quantum mechanical heat engine*, 2016
Høgni C. Kamban, Rasmus F. Andersen, Rune L. Riksted, *An examination of the absorption spectrum of MoS_2* , June 2016
Martin O. Mikkelsen, Jannick K. Jørgensen, Mads-Peter V. Christiansen, *Modeling of sodium clusters using density functional theory*, June 2016

Teaching

The QUSCOPE PIs teach relevant courses in their respective departments, and as part of their training, some QUSCOPE postdocs have run quantum optics and quantum information courses and contributed to the supervision of bachelor students. PhD students teach undergraduate exercise classes and have also run topical study group courses for their fellow students.

Outreach

The QUSCOPE research topics have high appeal to the general public, and members of the Centre have fostered many outreach activities including presentations to high school students and teachers.

More “exotic” outreach activities include the computer and smartphone game “Quantum Moves” and public lectures combined with cultural events, e.g., at Aarhus Theatre, the cultural venue “Godsbanen”, and Aarhus Festuge. Two “quantum music” projects with a classical composer feature scientific presentations along with a piano duo performance in Belgrade, November 2016, and with a concert for sinfonietta and cello, planned for The Royal Theatre of Copenhagen and the new Aarhus public library, March 2017.

Research network and international collaboration

The QUSCOPE members contribute proposals and analyses that are being used by a number of experimental groups and we collaborate with a large number of scientists worldwide. Here we list some of our more significant long term collaborations.

- Daniel Esteve and Patrice Bertet, Quantronics Group, CEA (French Atomic Energy Commission), Saclay, France - 6 joint publications with Klaus Mølmer
- Mark Saffman, University of Wisconsin, Madison, USA - joint grants and a joint publication with Klaus Mølmer
- Kater Murch, Washington University, St. Louis, Missouri, USA - 3 joint publications with Klaus Mølmer
- David Petrosyan, Institute of Electronic Structure and Laser, FORTH, Crete, Greece - 6 joint publications with Klaus Mølmer and co-supervision of postdoc Lukas Buchmann.
- Oleg Tolstikhin, Moscow Institute of Physics and Technology, Moscow, Russia – 2 joint publications with Lars Bojer Madsen and co-supervision of PhD student, Jens Svensmark.
- Tony Starace, University of Nebraska, Lincoln, Nebraska, USA - 2 joint publications with Lars Bojer Madsen
- R. Wörner, ETH Zürich, Switzerland – 3 joint publications with Lars Bojer Madsen
- B.K. Nikolic, Department of Physics and Astronomy, University of Delaware, Newark, Delaware, USA - 4 joint publications with Thomas Garm Pedersen
- A. Harju, COMP Centre of Excellence, Department of Applied Physics, Aalto University, Helsinki, Finland - 1 joint publication with Thomas Garm Pedersen
- V.M. Pereira, Centre for Advanced 2D Materials, National University of Singapore, Singapore - 2 joint publications with Thomas Garm Pedersen
- V. Veniard, Laboratoire des Solides Irradiés, Ecole polytechnique, CNRS, CEA, Université Paris-Saclay and European Theoretical Spectroscopy Facility, Palaiseau, France - 1 joint publication with Thomas Garm Pedersen

Additional external funding

Lars Bojer Madsen

2011-2016 ERC Starting Grant: € 1.33M

2014-2018 PI in the ITN-ETN Marie Skłodowska-Curie MEDEA: € 290,000

Thomas Garm Pedersen

2012-2017 “Center for Nanostructured Graphene”, Center of Excellence grant from Danish National Research Foundation, share 1/9 of 54MDkr

Klaus Mølmer

2013-2016 FNU project, Many-particle interference, 2.199.550 DKK

2011-2015 EU-ITN COHERENCE, 1.710.082 DKK

2015-2018 EU H2020 RySQ, 1.695.294 DKK

2015-2020 US Army Research Laboratory, Collaborative grant CDQI, 3.465.000 DKK

2016-2019 EU H2020 NanOQTech, 2.974.000 DKK

2012-2022 Villumfondens Aarslegat, 2.000.000 DKK

The Research Minister’s EliteForsk Travel Award to PhD student Christian Kraglund Andersen, January 2014-July 2016, 200.000 DKK

The Research Minister’s EliteForsk Travel Award to PhD student Alexander Holm Kiilerich, January 2016-July 2018, 200.000 DKK

Research

Background

Rather than organizing our scientific goals by the QUSCOPE research areas *quantum optics*, *ultrafast laser science*, and *nanooptics*, we identify our activities with three themes which transcend the conventional topic borders, and which to a larger extent reflect the current international developments in quantum research.

A. Control of quantum system dynamics with light

Laser light drives dynamics in quantum systems, and allows atomic states to function as bits in a quantum computer, strong fields to ionize atoms and break up molecules in selected final products, and excitations to enter and propagate inside and at interfaces of solid state systems. While these processes and their applications differ significantly in different systems, it is a common challenge that the evolution of a quantum system has to be effectively described and the most important processes identified under the combined action of laser driving, internal few- and many-body interactions, and damping mechanisms.

B. Generation of light by quantum systems

QUSCOPE investigates processes in which an external light source, typically a laser, drives a quantum system and forces the system to emit radiation with different frequencies and with non-classical properties. We provide theory for the quantum sources and for the application of the radiation generated and for the prospect to radiatively couple different quantum systems, e.g., for quantum (and classical) information processing.

C. Probing and precision sensing with light and quantum systems.

Ultrashort, ultraintense laser pulses permit investigations of electron and nuclear motion in molecular systems and promise major breakthroughs in our understanding of many- electron quantum dynamics in atoms, molecules and solids. In measurements on a single quantum system, “God plays dice” and theory is developed to describe the behavior of the observed system.

After presentation of some of our research highlights, we provide here a status of our research organized according to the work packages and research milestones. [Numbers in brackets] refer to the list of publications in the end of the report.

Research highlights

Highlight on time-dependent many-electron theory: The development of light sources with durations from the femtosecond to the attosecond timescale and with frequencies spanning from the mid-infrared to the XUV regime, offers the opportunity to investigate quantum systems on their natural time and length scales. We have developed an accurate description of many-electron systems, including an account for the electron-electron interaction in a fully time-dependent framework. Our theory is based on active space concepts borrowed from quantum chemistry, and elucidates, among other things, the role of electron-electron correlation in the process of high-order harmonic generation and the phenomena of enhanced ionization. See WP A.1.

Highlight on hypergeometric resummation: Several problems in quantum physics have a non-perturbative character. Hence, one cannot rely on gradually adding more and more terms to an approximate initial solution. An important example is the probability of ionization of a bound quantum state by a strong electric field. We have developed a novel approach that tackles a large class of these problems ranging from ionization over non-perturbative energy shifts to influence of nuclear motion on electric currents. The approach is based on “hypergeometric resummation”, which allows summing infinitely many contributions to a particular phenomenon in a simple manner. This versatile tool has subsequently resulted in 4 publications in prestigious scientific journals. See WP A.2.

Highlight on hybrid quantum technologies: We have worked on new designs for super conducting electronic circuits and on protocols to interface them with long lived atomic degrees of freedom. Our scheme for quantum state transfer from a Josephson junction qubit to an atomic ensemble has been successfully demonstrated, and we have shown that the same physical set-up can be applied as a microwave sensor, a million times more sensitive than previous spin spectrometers. See WP A.4.

Highlight on nonlinear many-body response: The behaviour of many-body quantum systems under external influences such as optical and electric fields is a challenging task. Even isolated many-body systems are themselves complicated objects that are difficult to model. Including external disturbances add another layer to the complexity. We have studied linear and nonlinear responses of such systems to static and dynamic electric fields. This has enabled simulations of light-emission, photodetection and frequency conversion in various quantum systems. Moreover, in collaboration with experimental partners, the nonlinear response of the semiconductor MoS₂ have been given a satisfactory explanation. See WP B.2.

Highlight on Attosecond Physics: In many chemical reactions, biological processes and technical applications, the ultrafast transfer of charge plays a very important role. In addition to the driving of such processes by nuclear motion, there is a much faster solely electronic response known as charge migration. Charge migration occurs on the attosecond time scale and involves the coherent concerted motion of electrons. Using state-of-the-art experimental techniques and combinations of advanced quantum chemistry and theoretical physics models, the attosecond charge migration in iodoacetylene was measured and controlled. See WP C.1.

Highlight on quantum trajectories: Systems subject to probing undergo stochastic trajectory dynamics according to the random outcome of measurements. In a series of works, we have demonstrated the connection to precision measurements and how the theory can be extended to also retrodict the earlier state of a system and the strength of a physical perturbation. The analysis has been applied with striking results on cavity QED and superconducting qubit experiments, and we currently work on extensions to quantum dots, nuclear spin dopants and optomechanics where the retrodiction provides crucial dynamical information. See WP C.5, and C.6.

Work package and milestone reports

On the following pages we review our progress within all work packages. For convenience, we recall the Year 2 and Year 5 milestones, envisioned for each work package at the beginning of the project.

Theme A: Quantum dynamics induced and controlled by light

Workpackage A.1: Attosecond transient excitation of electronic and nuclear motion in atoms, molecules and solids

Year 2 milestones:

- Implementation of TD-RASCI and TD-RASSCF for 3D systems
- Simulation of ionization (and possibly transient absorption spectroscopy experiments) in atoms and molecules under attosecond light transients

Year 5 milestone:

- Studies of transient absorption in condensed matter systems

The implementation of the TD-RASCI and TD-RASSCF theories for 3D systems was finalized in 2016, and the methodologies are currently being applied to a range of problems. The TD-RASCI theory was extended to a more general treatment of the active space and this extension is named TD-generalized-active-space-CI (TD-GASCI) [12,52,112], and two manuscripts J. J. Omiste, W. Liang Li and Lars Bojer Madsen, “The three-dimensional time-dependent restricted-active-space self-consistent-field method: Application to beryllium”; Haruhide Miyagi and Lars Bojer Madsen, “Time-dependent restricted-active-space self-consistent-field theory with space partition” are almost ready for submission.

Transient absorption spectroscopy has been studied in Refs. [31,41,73,89]. In particular molecular effects associated with nuclear motion and orientation of the molecule in attosecond transient absorption spectroscopy (ATAS) has been elucidated. Figure A.1 shows an example in molecular

hydrogen. The figure is highlighting the difference between the results for the spectra with fixed and moving nuclei.

In the condensed matter phase, studies have been initiated on high-order-harmonic generation in gapped graphene with femtosecond pulses [110].

In the remaining duration of the center, work on ATAS will continue. Ongoing and future work involves analytical approaches to different features in the ATAS spectrum based on adiabatic theory, effects of non-Born adiabatic coupling in molecules, including investigation of the role of conical intersections. As seen from the above, the focus in this workpackage has shifted from the original year 5 milestone involving ATAS in condensed matter systems to studies in atoms and molecules.

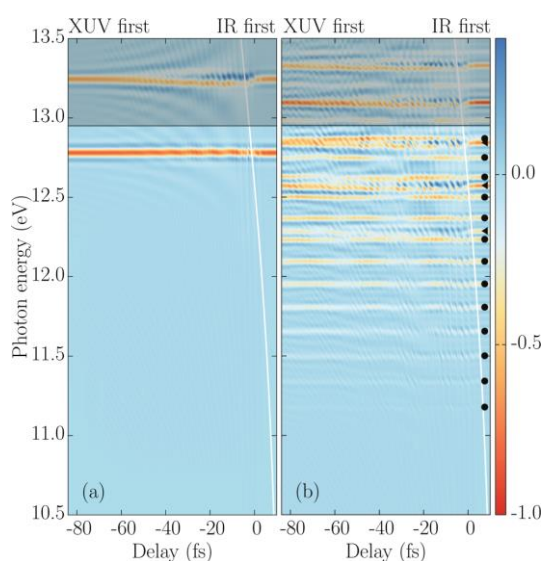


Figure A.1: Normalized ATAS spectrum for molecular hydrogen for (a) fixed nuclei and (b) moving nuclei [31].

Workpackage A.2: Correlated electron-nuclear motion

Year 2 milestones:

- Detailed studies of energy sharing between electrons and nuclei in diatomic molecules
- Nuclear motion in tunneling in heteronuclear systems in model systems and analysis of the breakdown of the BO approximation in the weak-field regime

Year 5 milestones:

- Elucidation of energy sharing in condensed matter systems
- Nuclear motion in tunneling in 3D

Detailed studies of the energy sharing between electrons and nuclei in diatomic molecules subject to time-dependent laser pulses have been reported in Refs. [11,38,60,111]. Figure A.2 is an example from [60] showing the joint energy spectrum of electrons and nuclei after laser-induced dissociative ionization of the hydrogen molecular ion. The figure highlights how the energy sharing is affected by changing wavelengths.

Effects of nuclear motion on tunneling ionization, including analysis of the breakdown of the Born-Oppenheimer approximation, and derivations of weak-field asymptotic theory were reported in Refs. [15,71].

Nuclear motion (phonons) also affects electron transport in condensed matter systems. In a diagrammatic approach, the electron-phonon interaction can be included up to a certain order. We have applied the hypergeometric resummation approach to effectively approach infinite order within the self-consistent Born-approximation [90].

In molecular systems, the physics and effects of nuclear motion are phenomenologically well understood based on 1D modeling. The implications of 3D nuclear motion have been discussed in the literature. For these reasons, the former year 5 milestone on 3D nuclear motion does no longer have high priority.

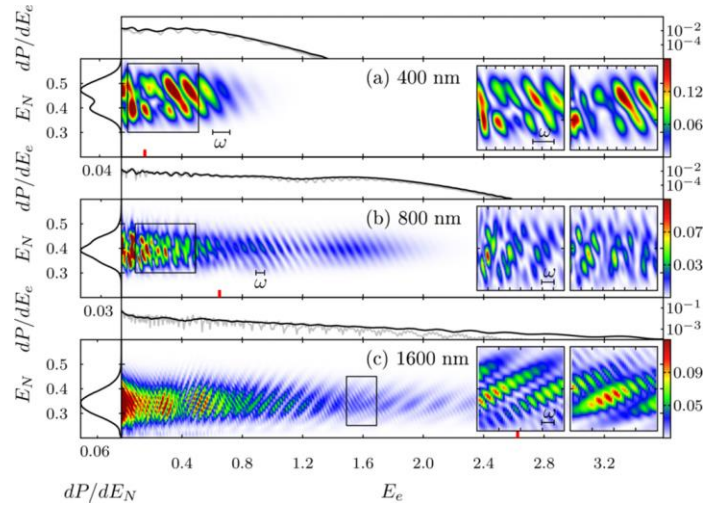


Figure A.2: Joint energy spectrum of the hydrogen molecular ion after expose to intense ($3 \times 10^{14} \text{ W/cm}^2$), 6 cycle pulses with the wavelength indicated in the panel. See Ref. [60] for details.

Work package A.3: Quantum information with neutral atoms

Year 2 milestones:

- Numerical methods and analyses of excitation dynamics in a single atomic cloud and in a collection of weakly coupled clouds of strongly interacting Rydberg atoms.
- New proposals for implementation of quantum computing gates in the presence of dissipation and dephasing.

Year 5 milestones:

- A consistent understanding of the interplay between unitary and dissipative dynamics in few-atom and multi-atom systems.
- Schemes for time dependent gate dynamics driven or assisted by dissipation and dephasing.

We have met the Year 2 milestones by developing and investigating the performance of a number of new schemes [8,19,66] for quantum information processing with neutral, Rydberg interacting atoms. While dissipation and loss is treated as a source of error in the above analyses, we have also proposed schemes that effectively *rely on* dissipation and decay to prepare atomic ensembles in

particular ordered or entangled quantum states [13,30,63]. These and our more recent works [92,95,113] bring promise that we shall successfully accomplish also our Year 5 milestones.

In [92], carried out in collaboration with experimentalists in Hefei, China, we employ the so-called Rydberg blockade effect which allows an atomic ensemble to absorb a single collectively shared excitation. Following earlier Arhus theory proposals, excitations can be induced in different internal states of the atoms, and they can be subsequently released and detected as single photons. The system allows demonstration of the famous Hong-Ou-Mandel effect: coherent mixing of two single excitation states yields only states with two and zero quanta in the output modes. In our system, the correlations are verified by the statistics of the emitted optical photons, see Fig.A.3.

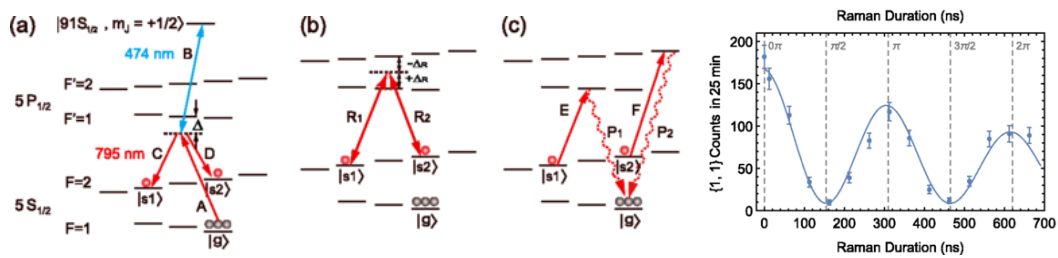


Figure A.3: Experimental procedure in [92]: (a) the ensemble is excited from $|g\rangle$ towards high-lying ($n=91$) Rydberg states and onwards to hyperfine states $|s1\rangle$ and $|s2\rangle$. (b) The hyperfine states are coherently coupled. (c) Atomic population in $|s1\rangle$ and $|s2\rangle$ is transferred to optically excited $5P$ states, which decay by emission of fluorescence. The right panels shows coincident detection of a photon both from the $|s1\rangle$ and $|s2\rangle$ states) as function of the duration of the coupling pulse in panel (b); coincidences are absent (HOM effect) in the $\pi/2$ and $3\pi/2$ cases corresponding to even mixing of the modes.

The experiments further demonstrate a so called NOON-state with quantum enhanced sensitivity to external perturbations. Further collaboration with the Hefei team will explore multi-mode, multi-excitation dynamics in Rydberg media.

Workpackage A.4: Quantum information with hybrid, atomic and solid state systems

Year 2 milestones:

- Quantum theory of coupled (hybrid) quantum systems
- Canonical quantization of superconducting circuits coupled to fields and atomic systems

Year 5 milestones:

- Proposal and optimization of schemes for quantum state processing in hybrid solid state and atomic systems.
- Schemes for high resolution detection with hybrid quantum systems.

We have presented a comprehensive overview of the challenges and reviewed a series of solutions to couple and transfer quantum states among different physical systems [25]. Together with experimentalists as CEA, Saclay, France, we have demonstrated the first transfer of quantum states from short lived electronic circuit into long lived atomic spin ensembles [40,83].

As a spin-off of this research [61], we have realized that very few atomic spins can be sensed by our microwave signals, and with CEA we have demonstrated the use of our quantum memory protocol

as an ultra-sensitive electron spin spectrometer. Figure A.4 shows an echo signal from just few hundred spins, an improvement in sensitivity over previous spectrometers by a factor 10^6 .

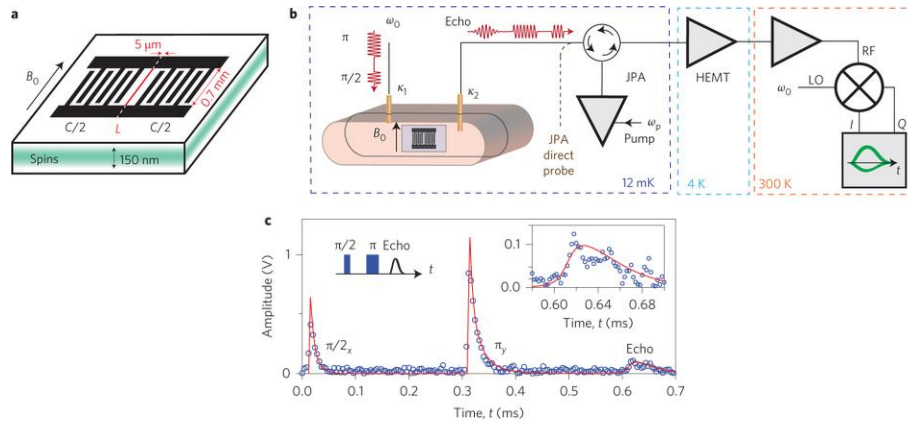


Figure A.4: (a) A spin ensemble, doped inside a crystal, is probed [61] via a microwave signal reflected from a superconducting circuit system that interacts weakly with the spins. (b) The signal is amplified in several stages from the 12 mK sample and finally read out as a classical current. (c) In the experiment, the spins are excited at time $t=0$, hit by a strong refocusing pulse after 0.3 ms , and they emit an echo signal around $t=0.6 \text{ ms}$. The red curve is our theoretically calculated signal, in good agreement with the measured signal (blue spheres).

We have recently submitted a theory analysis reaching single spin sensitivity [103] and a joint work with experimentalists [106] demonstrating the first use of quantum squeezed microwaves for precision measurements and are thus well underway towards our year 5 milestones.

Workpackage B.1: Nonlinear optics; single active electron models

Year 2 milestones:

- TD-RASCI single-active-electron results for HHG processes in atoms and small molecules
- Perturbation calculation of nonlinear optical processes in graphene and carbon nanotubes

Year 5 milestone:

- Full non-perturbative calculation of HHG in solid-state structures at the single electron level

The TD-GASCI single-active electron methodology was formulated as part of a detailed description of the theory [12]. The methodology was applied to the study of enhanced ionization in diatomic molecules, where the role of electron-electron correlation was elucidated by comparing the results of calculations for this process at different levels of theory (TDHF, TD-GASCI-singles, etc.) [52]. More work in this direction is in progress. It was judged that the process of enhanced ionization was more interesting for applications of our theory than the process of HHG, where already a range of studies with, e.g., TDCI-singles and R-matrix approaches, are available in the literature.

Calculations of the perturbative generation of low-order harmonics in graphene have been performed [110]. The extension to a non-perturbative description of HHG in graphene has been initiated in a two-band model [110].

The nonlinear optical response of solid-state systems has been modelled at the independent particle level. Focusing on two-dimensional semiconductors such as biased bilayer graphene, we have studied frequency conversion [35, 50] and nonlinear photocurrent generation [80]. The intricate

dependence on external parameters such as gating and doping has been investigated, which has led to identification of resonance conditions for efficient nonlinear processes, see Fig.B.1.

The results in the direction of the year 5 milestone thus appear promising.

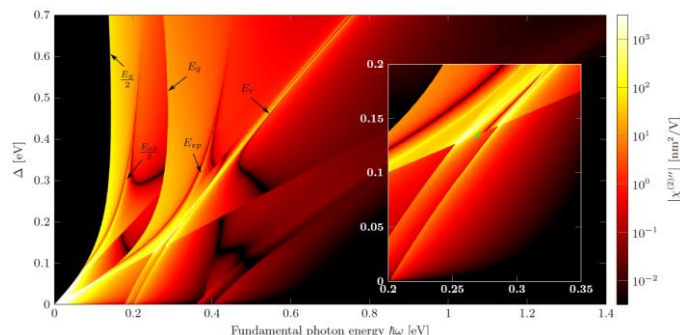


Figure B.1: The second-order nonlinear optical response of bilayer graphene as modulated by a perpendicular gate Δ [35].

Workpackage B.2: Nonlinear optics; multi-electron/correlation

Year 2 milestones:

- Many-body TD-RASCI and TD-RASSCF results for HHG processes in atoms and small molecules
- Studies of perturbative many-body effects in low-dimensional solid-state systems

Year 5 milestone:

- Non-perturbative many-body simulations of HHG in solid-state structures including graphene and carbon nanotubes

The TD-RASSCF theory has been used for studying HHG in atoms in 1D model studies [4]. The extension to 3D and molecules has involved several postdoc months, and the computer codes are being finalized and improved at present. In the remaining period of the center, these codes will be applied to a range of timely problems in strong-field and attosecond processes in atoms and molecules, not only involving HHG. In reduced dimensionality models of He and H₂ effects of electron correlation have been investigated in nonsequential double recombination in HHG, and a new molecular effect named same-period emission and recombination was identified [72].

Many-body effects in the nonlinear optical response of solid-state structures have been an important QUSCOPE area of research. The modelling has been done at the Bethe-Salpeter [50,53,80] and Mott-Wannier [78,85] levels. This has allowed us to explain experimental data for frequency conversion in MoS₂ [53]. Moreover, the operation of photodetectors can be analyzed and proposals for device optimization have been made. These results have recently led to collaboration with one of the leading experimental groups in the field at ICFO in Barcelona. These results have all been based on perturbative expansions to low order. However, a non-perturbative formalism has now been constructed [50] and it is planned to explore these phenomena in the immediate future.

Workpackage B.3: Plasmon enhanced matter-light interactions

Year 2 milestones:

- Detailed modeling of field-enhancement in plasmonic structures
- Investigations of plasmon enhanced nonlinear optical processes involving atomic targets

Year 5 milestone:

- Modeling of all-solid-state targets for plasmon enhanced ultrashort wavelength light sources

Following recent experimental progress, we have developed a general density matrix theory for plasmonic lasing by excitation of molecular emitters near metallic nano-particles, see Fig. B.3. Our theory allows derivation of, e.g., the plasmon number distribution and phase fluctuations [75], and two further publications on multi-level emitters and on spatially random distributions of emitter distribution are in preparation.

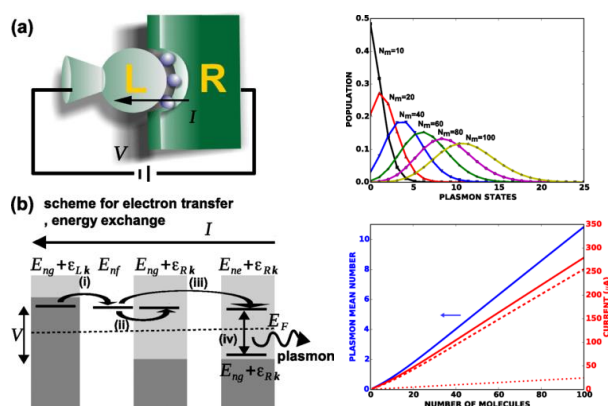


Figure B.3: The left panels sketch the physical system with molecules coupled to lead electrodes (the left spherical electrode supports a plasmon mode), which causes charging of the molecules in excited states, releasing their energy to the plasmon mode. The right upper panel shows the plasmon number distribution calculated in [75], and the lower panel shows the mean plasmon number (blue) as well as the current through the device (red solid) line. The dashed lines show contributions to the current proportional to the number of molecules (upper dashed) and to the number of plasmon excitations (lower dashed curve), respectively.

We have currently begun investigation of metallic waveguides and the potential to observe collectively enhanced coupling of emitters to wave-guided plasmon modes. New experiments with rare-earth ion doped nanocrystals have revealed excellent coherence properties, and plasmon modes may be used to couple such systems for quantum information tasks.

Workpackage B.4: Cavity QED and near field quantum optics

Year 2 milestones:

- Description of the quantum states of fields emitted by few and many-atoms.
- Theory for collective emission of light by microscopic quantum emitters, in the vicinity of passive dielectric components, and in the vicinity of metal surfaces.
- Quantization of superconducting circuit elements and their emitted radiations fields.

Year 5 milestones:

- Proposals for physical components that emit, detect, and manipulate quantum states of radiation in new ways.
- Study of phase transitions in coupled matter-radiation systems.

A recent result [95] suggests how single atom qubits can be coupled effectively to a single photon, by placing the former inside a cloud of atoms of a different species. An ensemble with N atoms thus amplifies the effective light-matter interaction by \sqrt{N} and further allows communication between atoms and photons at completely different wavelengths. Figure B.4 shows that the protocol achieves an optimal infidelity (error), $(1-F)$ scaling as $1/\sqrt{N}$.

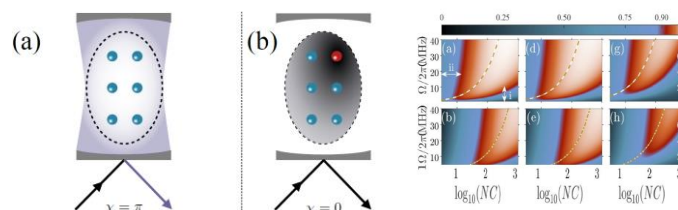


Figure B.4: Left panels: Schematic setup of individual atoms (balls) immersed in a cloud of different atoms (oval). The ground state atoms (blue) do not affect the cloud, but an excited atom (red) modifies the interaction of the cloud with incident photons. The right most panel shows the fidelity (ideally $F=1$) of a quantum phase gate between a single photon and a single atom (top row) and between two atoms mediated by a photon (bottom row) as function of atomic and field parameters of the scheme. The three columns represent different atomic level schemes.

In addition to optical cavities [95], we have studied coupling of atomic ensembles to mechanical oscillators [93], and to plasmons cf. Workpackage B.3.

In [23], we have devised a method to couple two quantum emitters with different frequencies to the near field of a waveguide for microwaves (with multi-sideband eigenmodes imposed by modulation of an inline SQUID) - a procedure which is now applied, in slightly altered form, in the IBM quantum computer.

Waveguide resonators are essential components that permit coupling between different systems, and we have provided [94] a general method to quantize systems consisting of both discrete circuit elements such as capacitors, inductors, and Josephson junctions, and of waveguides, whose quantum degrees of freedom obey continuous wave equations, with boundary values set by the discrete elements. These works meet the first milestones, while our efforts on superconducting circuits reported in Refs.[20, 23, 99, 101, 105] are bi steps towards the Year 5 milestones.

Workpackage B.5: Propagating light and cascaded quantum systems

Year 2 milestones:

- Theory for quantum systems driven by quantum light
- Analyses of non-linear processes as witnesses of the quantumness of driving fields

Year 5 milestones:

- Spectral and temporal effects on the propagation of quantum fields.
- Quantum effects in photon absorption from optical parameter oscillator sources and correlated frequency combs.

Our work on propagating light and cascaded quantum systems has led to contributions in microwave spectroscopy, where we have proposed and analyzed the use of cascaded Josephson Parametric Amplifiers to squeeze a probe field and to amplify signals in a noiseless manner after weak interaction with a medium [61,106], see also Workpackage A.4.

Of a more general theoretical nature, we have investigated how quantum interference can be characterized beyond single particle wave phenomena. We have developed theories for how an interferometric set-up, subject to arbitrary numbers of quanta in the different input ports yields coincidence count signals in the output ports. Submitting an array of beam splitters to single photons in all input ports yields output states that are exponentially hard to calculate, and we have presented strong criteria to test the accuracy of physical devices that solve this so-called boson-sampling problem [3, 21]. We have also identified how decoherence mechanisms and distinguishability between wave components yield similar degradation of interference signal for single particles, but differ significantly for higher coincidence signals [18, 21, 107], see Fig.B.5, and how entanglement manifests itself in systems of many particles [64].

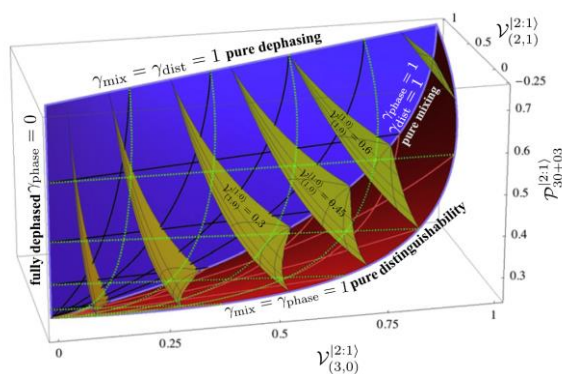


Figure B.5: In an imperfect interferometer, is the reduced fringe contrast due to one interferometer arm being subject to a random phase, a distortion of the wave reducing its overlap with the other arm, or entanglement with a local environment? The figure from [18] shows how three measurable coincidence properties, are sufficient to quantify how much of each of these three mechanisms are at play in a given set-up. The scheme uses three-photon input states, and an experimental group in Pohang, Korea, has made the first demonstration of such experiments (unpublished).

Our work on many-body interference led us to study more fundamentally the quantum statistics of identical particles and examine to what extent composite objects are bosonic or fermionic in nature. A diatomic molecule, composed of two fermionic atoms forms a bosonic particle, unless it is being probed at very fine detail, e.g., at close distance or high energy. We have shown that the essential quantity determining the bosonic or fermionic character of a bipartite system is the entanglement between its constituents and that the Pauli principle of its constituents modifies the bosonic commutator relations of the composite particle. The modifications are normally infinitesimal, but we have developed schemes that amplify their consequences in multi-particle multi-mode interferometers, where they may be detected [96].

Workpackage C.1: Ultrafast phenomena; *Atoms and Molecules*

Year 2 milestones:

- 3D Implementation of the single-active electron approximation in the TD-RASCI scheme and calculations addressing experimental data for lithium

- Angular distribution from double excited states of helium and analysis of electron-electron correlations effects
- Formulation of the weak-field asymptotic theory including multi-electron effects and validation in small system where comparison with *ab initio* results are possible (helium, lithium and the hydrogen molecule)
- New propagation scheme based on an adiabatic basis approach

Year 5 milestones:

- Tunneling theory including many-electrons: Extraction of relevant information from quantum chemistry calculations
- Attosecond streaking from condensed matter systems

We found that the year 2 milestones were too specific, and focus shifted to other processes within the same general topic. Specifically, in collaboration with Prof Starace, University of Nebraska, Lincoln, Nebraska, USA, electron vortices in photoionization of helium by circularly polarized attosecond pulses were studied [43,77], see Figure C.1. In collaboration with the experimental group of Prof Wörner, ETH, Zürich, Switzerland, laser-induced electronic structure in oriented polyatomic molecules [32], charge migration in ionized iodoacetylene [47], and influence of multi-electron dynamics in the asymmetry of strong-field ionization and fragmentation of the polar molecules in the methyl halide series were studied [51]. In a collaboration involving several leading laboratories, a new semiclassical theory for the two-step model was formulated [79]. The physics of low-energy electron following strong-field ionization was analyzed theoretically [27], and in collaboration with the experimental group of Prof Stapelfeldt, Aarhus University, Aarhus, Denmark [7,35], where also photoelectron momentum distributions [46] and alignment dependent ionization yields were analyzed [87]; see also [82] for a theory study. The use of bicircular fields for the generation of circularly polarized harmonics was considered from a rotating frame perspective [68], and it was pointed out how bicircular HHG can be used to illuminate the nuclear configuration within a molecule [86]. Finally the channel-resolved spectra in He were considered under intense short XUV radiation [69], and the extraction of Stark shifts of multiple states in the ion was pointed out [109].

The weak-field asymptotic theory was formulated including multi-electron effects. The formulation involves the Dyson orbital. At present the TD-GASCI method is used to include correlation in the ground state of H₂ and LiH, and to extract a correlated Dyson orbital from which the tunnelling rate can be determined.

Work has been initiated based on an integral formulation of the tunnelling rate. In a mean-field HF picture, this approach involves one- and two-electron integrals of the orbital from which tunneling ionization occurs with all the other orbitals forming the configuration of the ground state. Since the method is based on integrals and not asymptotic form of a wave function, it can be evaluated using standard quantum chemistry methods. Work based on the HF level of theory is in progress. Extension to methodologies including electron correlation seems possible. In the above sense, the year 5 milestone will be addressed.

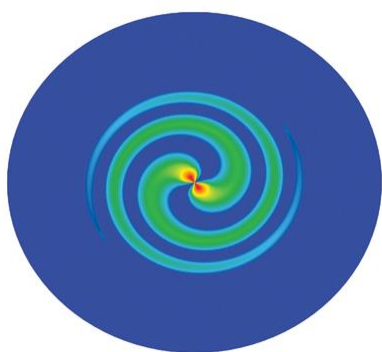


Figure C.1: Interference between photoelectron wavepackets in momentum space produced in the ionization of a helium atom by a pair of oppositely circularly polarized time delayed attosecond pulses [43]. Also on the cover of Phys. Rev. Lett. 115, and highlighted in Nature Physics 11, 800 (2015) as ‘Vortex mixer’.

Workpackage C.2: Ultrafast phenomena; *Condensed matter systems*

Year 2 milestones:

- 3D Implementation of the single-active electron approximation in the TD-RASCI scheme and calculations in lithium addressing unexplained experimental data
- Angular distribution from double excited states of helium and analysis of electron-electron correlations effects
- Formulation of the weak-field asymptotic theory including multi-electron effects and validation in small system where comparison with *ab initio* results are possible (helium, lithium and the hydrogen molecule)
- New propagation scheme based on an adiabatic basis approach

Year 5 milestones:

- Tunneling theory including many-electrons: Extraction of relevant information from quantum chemistry calculations
- Attosecond streaking from condensed matter systems

The traditional approach to optical response theory in condensed matter is based on frequency domain. However, to study ultrafast phenomena such as excitation and emission of ultrashort light pulses, it is advantageous to work in time domain. For this reason, we have initiated perturbative and non-perturbative studies of pulse excitation in 2D semiconductors. As a specific application, HHG has been studied in gapped graphene [110] including aspects of non-perturbative driving. In particular the change in a given harmonic when increasing the intensity and going from the perturbative to the nonperturbative regime was investigated; see Fig. C.2. The year 5 milestone on nonperturbative many-body simulations on HHG in solid-state structures will be addressed by including excitons in a density-matrix based formulation of nonperturbative dynamic response. This has already been initiated in the perturbative regime in Ref. [50].

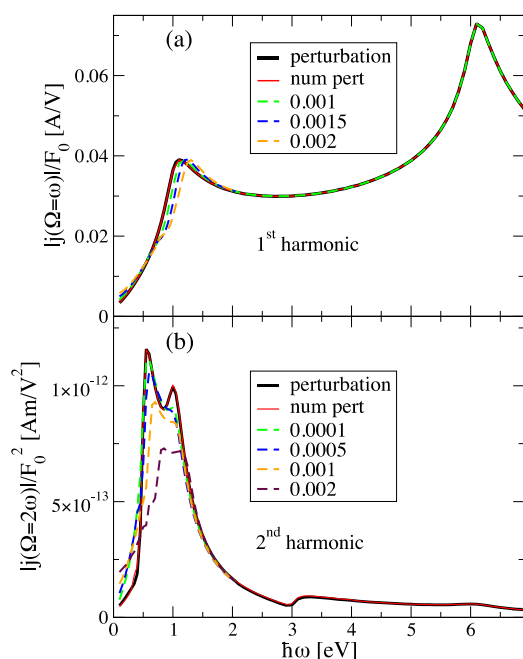


Figure C.2: Absolute value of the (a) first, and (b) second harmonic for gapped graphene subject to laser pulses with intensities ranging from the perturbative to the nonperturbative regime. See [110] for details.

Workpackage C.3: Magneto-optical spectroscopy

Year 2 milestones:

- Magnetic effects in excited states at the single-electron level including spin-orbit coupling.
- Single-electron models of the magneto-optical response of atomic, molecular and condensed matter systems.

Year 5 milestones:

- Magneto-excitons in low-dimensional condensed matter.
- Many-body effects implemented in both TDDFT and BSE approaches to magneto-optics.

The inclusion of magnetic fields in atomistic condensed matter models is challenging due to the breaking of translational invariance by the field. Hence, new computational tools are needed to treat realistic scenarios. As an initial step, we have considered the effects of magnetic fields in gapped and nanostructured graphene [81]. This has allowed us to fully incorporate magnetic effects at the single-electron level, c.f. year 2 milestones. So far, however, our focus has been on electron transport rather than optical response. Hence, as the next step, attention will be shifted to magneto-optical properties. These activities will be the focus of PhD program that has only recently started up (Aug. 2016). Moreover, the first calculations of Wannier-Mott magneto-excitons in transition-metal dichalcogenides have been made.

Workpackage C.4: Conditional quantum dynamics, heralding and feedback

Year 2 milestone:

- Propose practical means for entanglement generation and quantum computer gate operations by means of, or assisted by, measurement in diverse physical systems.

Year 5 milestone:

- Study generic criteria for feasibility and optimality of quantum state control by combined measurements and interactions.

Measurements in quantum mechanics are accompanied by so-called back action effects, where the state of the observed system changes according to the random measurement outcome. This provides a means to steer the dynamics and prepare desirable states of quantum systems that cannot be prepared by other means.

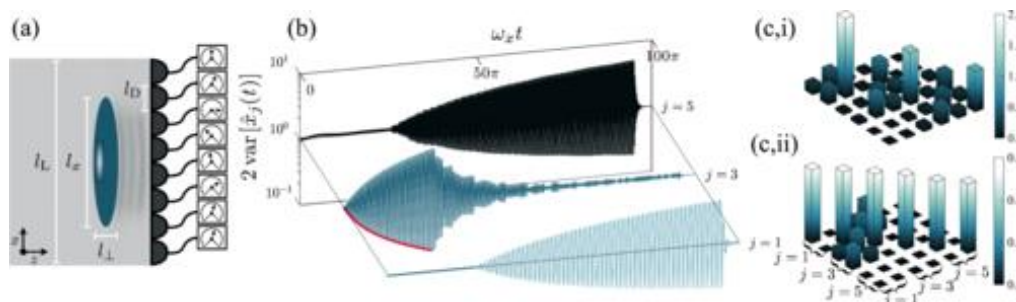


Figure C.4: (a) The array of detectors yield information about the spatial profile of condensate oscillations. (b) By varying the probe frequency, we address first the 3rd sound mode, which becomes squeezed, and later the 1st and 5th modes which become pairwise correlated (entangled). (c) Shows correlations between x and p quadratures of the modes 1,3 and 5; (c,i) for continuous probing shows correlations among all modes, (c,ii) for stroboscopic probing shows strong correlations between mode 1 and 5.

In [39,58], we have studied how off-resonant optical probing of a Bose Einstein Condensate, see Fig.C.4, reveals its density profile and hence establishes a quantum state of the sound modes of the system. Probing stroboscopically, we can address individual eigenmodes and squeeze their canonical quadrature components and entangle separate pairs of modes.

Measurements in conjunction with feedback permit a wider control of quantum systems, and will ultimately be needed to mitigate errors in quantum computing and communication architectures. We have developed detection schemes for the particular case of microwave circuits [99]. These systems have to be kept inside cryostats below 100 mK, and communication in and out of these systems is hampered by the need for thermal isolation and attenuation. We have thus proposed new autonomous protocols in which one microscopic system, inside a cryostat can perform measurements and automatically provide feedback on another one. This can be used to implement classical flip-flop gates at low power levels [20] and to prepare and correct quantum states [57].

Workpackage C.5: Quantum metrology; quantum Bayes inference

Year 2 milestones:

- Numerical methods for precision measurements based on stochastic master equations for photon counting, homodyne and heterodyne field detection.
- Studies of adaptive strategies to improve sensitivity by fine-tuning of measurement strategies according to previous outcomes.

Year 5 milestones:

- Fundamental limits of sensitivity based on (quantum) Fisher Information and Cramér-Rao bounds.
- Theory of optimal sensitivity of a controlled quantum system constrained by a set of available measurement strategies.

Measurement outcomes are random, and when “God plays dice”, the system is kicked, and we have demonstrated theoretically that by observing the transient dynamics after these kicks, and thereby inducing more kicks, we gain more information than is available in the average system behavior [2,16,17,45,84,103]. Our studies have led to the identification of the fundamental limits of continuous sensing, and they have provided practical means of analysis of the given experimental signals reaching the theoretical limits. Experimental partners at CEA, Saclay, France are now pursuing the proposal of Ref.[103], and we expect that they will be able to detect and manipulate a single electron spin dopant in a crystal.

Recently [108], we have addressed adaptive strategies, where the system parameters are changed according to earlier measurement outcomes. As we are not repeating a simple experiment, normal statistics does not apply, and we are faced with a complicated statistical analysis of the performance of specific strategies. An interesting scheme applies a random search, where a laser frequency is randomly shifted at each photon detection event, until it gets close to a “dark resonance”, where the fluorescence signal goes to zero. The behavior of this protocol is strictly non-ergodic, and will be dominated by the longest time interval with the closest proximity to the resonance, see Fig. C.5. This situation is explained by the laws of Levy-statistics [108], and we have obtained analytical expression for the accomplishments of the method. We believe that our new method of analysis can find applications for a wider class of adaptive schemes and protocols for parameters estimation and state control.

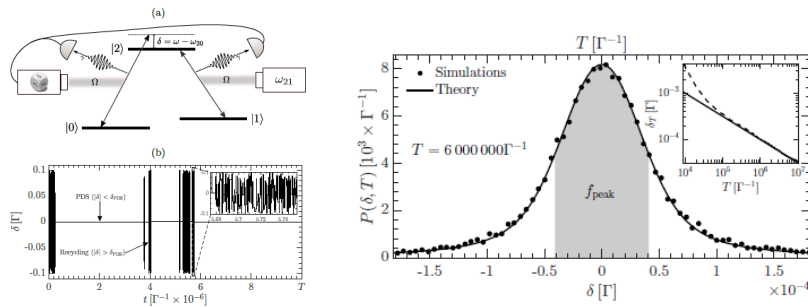


Figure C.5: Left (a) schematic of atomic level scheme and illumination by lasers that change frequencies when photons are detected; (b) simulated variation of the laser frequency, dominated by long periods where the laser is close to a dark resonance. Right: Simulated and analytical distributions of the frequency estimate based on the laser frequency attained at the end of the probing period (this value is likely to belong to one of the long dark period).

Workpackage C.6: Extension of quantum measurement theories: new challenges

Year 2 milestones:

- Develop past quantum state formalism for correlation functions and spectra.
- Proposals for experimental tests and applications of past quantum states.

Year 5 milestones:

- Application of quantum measurement theory to cascaded and concatenated quantum systems.
- Development of quantum measurement theory for new detectors based on phase transition and switching dynamics of superconducting circuit elements.

QUSCOPE has added new insight into the foundations of quantum optics by new analyses of the evolution of quantum systems, conditioned on the detection of their emitted radiation. In particular, we have developed a theory of “Past Quantum State”, which accounts for our knowledge about a

quantum system at time t , based on measurements carried out on the system, before and after t . The theory supplements the conventional density matrix, conditioned on previous measurement events with an additional matrix, representing posterior events - the “hindsight”, and like the normal applications of Born’s rule in quantum mechanics, we can retrodict the probabilities for outcomes of measurements, performed but not yet revealed to us.

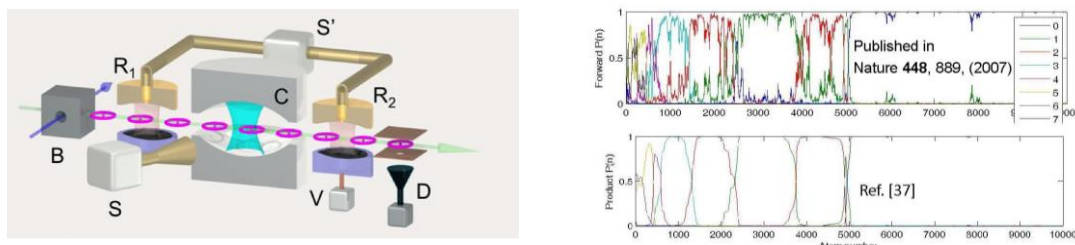


Figure C.6.: Left: Physical setup, where atoms passing through a cavity reveal partial information about the photon number in the cavity. Right, upper panel: Probabilities for different photon number states inferred from the atomic stream of data (Nature 2007). Right, lower panel: Significantly improved photon probabilities according to the past Quantum State formalism, which uses all atomic detection events to infer the photon number.

Applications of this theory range from collaboration with Nobel laureate Serge Haroche on the preparation and verification of photon number states in cavities [37], shown in Fig.C.6, to joint work with colleagues at Berkeley and St. Louis in the US on superconducting circuit dynamics subject to microwave probing [26, 67, 102]. The theory firmly establishes a connection between optical correlation functions and conditioned dynamics [44,48,].

Lately we have applied the formalism to optimize quantum teleportation protocols where the heralding measurements take finite time, and hence the optimal operation requires an estimate of “what was” rather than “what is” the state of a quantum system [91]. Three QUSCOPE PhD students currently use and develop the method in joint projects with experimental groups dealing with quantum dots, optomechanics and nuclear spin dopants, respectively.

Summary and outlook

In summary, the QUSCOPE Centre of Excellence has delivered a large volume of original research. In a few of our work packages, we report minor deviations from some of our original research plans and extensions of others. These have been motivated by new exciting possibilities offered by our theoretical discoveries and methods as well as by unique opportunities to apply our work in collaborations with world leading experimentalists and theorists.

The following two years will be an important period for QUSCOPE, where we shall promote and consolidate our major research results and we shall pursue new ambitious research goals through international collaborations. We shall continue to engage our talented students and postdocs in these collaborations, and we are determined to provide them with skills that will enable them to build their own research career and contribute to very active fields of research in science and technology.

Publications

At a glance, QUSCOPE published 13 articles in 2014, 42 articles in 2015, and 45 articles in 2016).

The QUSCOPE publications include 1 Nature, 1 Science, 1 Nature Nanotechnology, 2 Nature Communications, 1 PNAS, 15 Physical Review Letters, 14 Physical Review B, and 2 Invited News and Views articles in Nature and Nature Physics.

15 of our articles have been selected for special mentioning by editors of the scientific journals [indicated with ● ... ● in the publication list].

Full QUSCOPE list of publications

2014

1. P. Haikka and K. Mølmer, *Dissipative Landau-Zener level crossing subject to continuous measurement: Excitation despite decay*, Phys. Rev. A **89**, 052114 (2014).
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6. Klaus Mølmer, *Quantum memory: Needle in a haystack*, Nature Physics **10**, 707–708 | news & views (2014).
7. D. Dimitrovski, J. Maurer, H. Stapelfeldt, and L. B. Madsen, *Low-Energy Photoelectrons in Strong-Field Ionization by Laser Pulses with Large Ellipticity*, Phys.Rev.Lett. **113**, 103005 (2014).
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14. John J. L. Morton and Klaus Mølmer, *Quantum information: Spin memories in for the long haul*, Nature **517**, 153–154 | news & views (2015).
15. Jens Svensmark, Oleg I. Tolstikhin, and Lars Bojer Madsen, *Coulomb and dipole effects in tunneling ionization of molecules including nuclear motion*, Phys. Rev. A **91**, 013408 (2015). • PRA Kaleidoscope Images •
16. Alexander Holm Kiilerich and Klaus Mølmer, *Parameter estimation by multichannel photon counting*, Phys. Rev. A **91**, 012119 (2015).
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28. M. R. Thomsen, S. J. Brun, and T. G. Pedersen, *Stability and magnetization of free-standing and graphene-embedded iron membranes*, Phys. Rev. B **91**, 125439 (2015).
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