Quantum and Nonlinear Optics

Report of Abstracts

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Abstract ID : **1**

Stars and their Spectra. From historical beginnings to challenges of tomorrow

Content

\title{Stars and their Spectra. \\
From historical beginnings to challenges of tomorrow }
\author{Dainis Dravins (Lund Observatory)}
\date{}

\begin{document}
\maketitle

Detailed spectroscopy of astronomical objects started with Joseph von Fraunhofer's studies of solar spectral lines around 1810, although he did not yet interpret them in terms of physical conditions on the Sun. Some decades later, Gustav Kirchhoff and Robert Bunsen showed how different chemical elements cause emission and absorption lines. In the same epoch, new observatories were erected worldwide, and astronomers started to look at stellar spectra. The relation between precise spectral-line wavelengths and radial motion of the source became understood, and in the late 19th century, first measurements of such a Doppler effect in different stars were realized. From the laws of planetary motion, it followed that sufficiently precise measurements would reveal the presence of smaller bodies in orbits around stars. Following decades of developments, discovery of the first exoplanets was achieved toward the end of the 20th century. However, a major challenge still remains in finding planets similar to the Earth. The limitations no longer are the imprecision of instruments but rather the intrinsic variability of stellar surfaces which causes a jittering of spectral-line wavelengths much greater than the tiny signal expected from small Earth-like planets.

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Submitted by **DRAVINS, Dainis** on **Friday, February 25, 2022**

Abstract ID : 2

Participation in COST action MD-G

Content

Participation in COST action MD-GAS

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It is my pleasure to be as a representative of Latvia on the Management board of European Cooperation in Science and Technology (COST) action Nr 18212, Molecular Dynamics in the GAS phase (MD-GAS) starting at 12th November 2019, and ending at 11th November year 2023.[1] The project proposal was submitted from Stockholm University and chair of action is professor Henning Zettergreen [2].
COST is a funding agency for research and innovation networks. This Actions helps to connect research initiatives across Europe and enable scientists to grow their ideas by sharing them with their peers. This boosts their research, career and innovation.
The main aim of the Action is to develop a new physical and chemical toolbox to significantly advance the understanding of: Gas phase molecular dynamics induced in interactions between molecules or clusters and photons, electrons, or heavy particles. Its consequences for a broad range of applications in e.g. astrochemical and atmospheric sciences, and molecular radiation damage.
The work is organized in three following work groups: 1st New high-performance instrumentation and experimental methods, 2nd Survival and destruction of molecules following energetic processing, and 3rd Charge-, energy flow, and molecular growth in intermolecular and intracluster reactions.
Despite the fact that the name MD-GAS does not correspond well to the research directions of our laboratory, this Action is attractive for us because one working group is engaged in the development of instruments and the campaign tends to have practical applications in the physics and photochemistry of the atmosphere. And the access to the DESIREE [3] facility via action is promising for our future development.
In connection with that I already have close and productive contacts with the group in Stockholm's University, see for example [4].
In my presentation I'll describe more in details my activities in COST actions and future plans for common experiments on DESIREE facility.

Acknowledgments
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Abstract ID : 3

Laser probing of metastable Ba+ for lifetime measurements

Content

Laser probing of metastable Ba+ for lifetime measurements

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Metastable levels are responsible for parity forbidden lines occuring in many low-density astrophysical plasmas, found in e.g gaseous nebulae, planetary nebulae, protostars, stellar chromospheres. Line ratios from forbidden lines are the most reliable tools for diagnostics of temperatures and densitiy of these regions. Measurements of metastable lifetimes is of direct importance for the use of forbidden lines.
We propose to further develop the laser probing technique to measure lifetimes for stored positive ionsusing DESIREE. One of the prime ions, thanks to its favourable atomic structure, is Ba+. The experimental results from this kind of measurements will test atomic transition probability calculations for multipole radiation in complex ions. M1 transitions can often be calculated with high accuracy, wheras E2 transitions are more challenging.
In the general case, by combining the experimental lifetimes from DESIREE with astrophysical branching fractions of the forbidden lines, we will provide experimental data for forbidden lines.
Most excited levels in neutral and near-neutral atoms and ions have radiative lifetimes of nanoseconds. However, some low-excitation levels are prevented to decay through normal electric-dipole, E1, radiation and thus decay rather through higher multipole radiation E2 and M1. With these radiations having many orders of magnitude lower transition rates, the lifetimes are in the range ms to 100s. The light from the metastable levels appear as forbidden lines, dominating the spectra from low-density astrophysical
plasmas appearing in e.g. gaseous nebulae, planetary nebulae, protostars, stellar chromospheres but also inthe outflows from supernovae. The forbidden lines are the key diagnostic tools for these regions. To be able to use the forbidden lines for important diagnostics of the physical conditions, such as temperature, density, chemical abundance, the lifetime of the metastable level and the transition rates of the forbidden lines must be known.
The current proposal is thus an effort in the development of a laser induced fluorescence technique for DESIREE. One of the most favourable ions to develop the technique of laser probing of a stored ion beam is Ba+. The atomic structure is simple with few levels and the metastable energy levels are located at low excitation energies. This allows for a high population and increased fluorescence signal, making Ba+ an ideal target ion. Figure 1 shows the atomic structure with the metastable 5d 2D levels along with the
ground state 6s 2S and the higher 6p 2P. A laser with 586 or 614nm is tuned to the 5d-6p transition, which emties the 5d states to the 6p. The subsequent decay to 6s is monitored and is a measure of the relative population of the 5d levels. By applying the laser at different delay times after injection, the lifetime curve is built up. The technique is described below along with the improvement suggested for the next experiment. In a separate section the results from an earlier experiment week are described.
The laser probing technique (LPT) described in the current proposal was derived by Mannervik and his group at the CRYING storage ring, and successfully applied to a number of ions of varying complexity [1]. For several complex ions, the measured lifetimes were combined with astrophysical line ratios to derive experimental transition rates [Experimental Oscillator Strengths for Forbidden Lines in Complex Spectra, Hartman, H, Johansson, S, Lundberg, H, Lundin, P, Mannervik, S, Schef, P, Physica Scripta, Volume T119, pp. 40-44 (2005)].

The LPT utilizes a cw laser to probe the number of atoms in the metastable states as a function of time. An ion beam of the element studied is stored, and a small fraction of the ions are in metastable states (5d for Ba+). A laser tuned to a transition to a higher level is is applied to empty the studied state (5d) to an upper, shortlived level (6p). The prompt decay of the 6p level is montitored by a PMT, and this fluorescence intensity is a measure of the population of the 5d level. By varying the delay after injection,

Figure 1: Level diagram of Ba+ showing the metastable 5d 2D states targeted in the present proposal, the 6p states being excited by the LASER tuned to the 586 and 614 nm, respectively, and the fluorescence at 456 nm.

and thus allowing the metastable 5d level to decay, the lifetime curve is built up from subsequent injections. Corrections must be made to the collisional processes with the rest gas, the varying ion current between injections and intra-beam losses. The significant repopulation from collisions with the rest gas observed in CRYRING is not observed in the DESIREE data. The DESIREE facility allows other states and ions to be measured, and the current proposal is an effort to develop the LPT. Its different conditions to previous and other storage rings require a modified technique, and other corrections.
When the Ba+ ions are produced in the ion source, a small fraction of the ions are in the excited 5d levels.
In the preliminary analysis of previous data, we are limited by several factors due to the intra-beam scattering which can be overcome if the fraction of ions in the excited states compared to the ground state could be increased. The fluorescence signal would then be increased by orders of magnitude, whereas the corrections to beam loss through space charge would stay the same. One efficient way to increase the 5d population is through active population of the excited states using an additional laser, rather than have to rely on the population from the ion source. This would then be tuned to the transition 6s-6p. Since the majority of the ions initially are in the ground state, this will excite the 6p states with prompt, subsequent decay to the 5d states. The atoms returning to the ground state from the 6p states will be re-excited by subsequent laser pulses. Using this active laser population, timed to appear prior to the probe pulse, the excitation of the 5d, and thus the recorded signal, will increase with several orders of magnitude while the corrections depending on the ion currents are unaffected.
Another important parameter for an accurate lifetime determination is the ion current which measures the total number of ions; the ion current will be measured through a Schottky spectrum to allow for accurate ion currents and its variations.The expected lifetimes for the 5d 2D states are extremely long with 80 and 32 seconds, respectively.
Thanks to the excellent ion storage conditions in DESIREE this can still be measured.

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Abstract ID : 4

Isotope Shift Measurements on Electron Affinity for Cl-

Content

Isotope Shift Measurements on Electron Affinity for Cl-

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The specific mass shift is sensitive to electron correlation that is particularly pronounced in negative ions. Hence, a study of isotope shifts in electron affinities is an excellent method to obtain benchmark data for theoretical models that go beyond the independent-particle model.
The isotope shift between the stable chlorine isotopes 35Cl- and 37Cl- was the first isotope shift in a negative ion to be investigated both experimentally and theoretically by Berzinsh et al. [1]. In their work a discrepancy between the experimental and theoretical result was observed. In 2013, this discrepancy was solved through state of-the-art ab initio calculations by Carette and Godefroid [2] using non-relativistic many-body theory. It will be difficult to transpose these correlation models from the non-relativistic to the full relativistic multi-configuration schemes, but there is some hope to take advantage of the partitioned correlation function interaction approach [3]. In this context, a study of chlorine isotopes with a large mass difference and hence a larger isotope shift would be very valuable.
In the list of Cl isotopes [4], we find them from mass 28 to 52. From them 35Cl and 37Cl are stable, and 36Cl has 3.01×105 years long ½ lifetime. All others a short lived. From mass 34 to 38 they exhibit lifetimes of about ½ hour long. Such isotopes can be studied at the online facility ISODE at CERN. All other isotopes have shorter lifetimes from seconds to picoseconds, which decrease with increasing mass difference from stable isotopes 35Cl and 37Cl .
The traditional spectroscopic methods, which are used to identify and to determine the structure of atoms and positive ions, cannot be applied to negative ions, and this also applies to radioactive isotopes. We propose to conduct measurements of the isotope shift in the electron affinity (EA) for chlorine isotopes at CERN by using Multi Ion Reflection Apparatus for Collinear Laser Spectroscopy (MIRACLS) [5] for the long lived 36Cl isotope, and the Gothenburg ANion Detector for Affinity measurements by Laser PHotodetachment (GANDALPH) apparatus [6].
The experimental method and apparatures will be presented in our report.

Aknowledgements
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Abstract ID : 5

Investigation of nonlinear optical processes in quantum dots and nanoparticles

Content

Investigation of nonlinear optical processes in quantum dots and nanoparticles

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Abstract: We report the investigation of nonlinear optical processes such as third-harmonic generation, nonlinear refraction and nonlinear absorption in the family of mercury containing quantum dots (QD): HgTe, HgSe, HgS, PbS QDs and Bi2Te3 nanoparticles (NPs) using the laser pulses with nanosecond, picoseconds and femtosecond range pulse duration. Nonlinear optical response has been studied in the broad spectral region 400 – 1700 nm. High conversion efficiency (˜10-4) towards the third harmonic (TH) of the 900 - 1700 nm, 150 fs laser in the thin (70 - 200 nm) films containing QDs deposited on the glass substrates is obtained. We analyzed spectral dependencies of the TH, nonlinear refractive indices, and nonlinear absorption coefficients of QDs and NPs thin films.

The growing interest in quantum dots (QD) and exfoliated topological insulator nanoparticles (NP) is attributed, particularly, due to their large low-order optical nonlinearities, which can be useful in photonics and optoelectronic devices in different time scales [1-6]. QDs and NPs have properties intermediate between bulk semiconductors and discrete atoms or molecules. Their characteristics change as a function of both size and shape. For these reasons, novel synthesized semiconductor QDs and NPs require to be examined under different conditions using laser pulses of variable energies, wavelengths, and durations to understand the nonlinear optical mechanisms and distinguish the attractive properties of those processes for practical applications.
The impressive third-order nonlinear optical properties make nanoscale PbS potentially useful in electroluminescent devices like light-emitting diodes and optical devices such as optical switchers. Special attention to the thin films containing these QDs is also related to the growth of such parameters as the nonlinear refractive indices, nonlinear absorption coefficients, and saturation intensities. The optical nonlinearities of nanoscale semiconductors in the quantum confinement regime have been frequently reported in the literature. Quantum confinement is found to enhance the nonlinear optical response, and a number of reports have discussed the physical processes underlying the mechanism of optical nonlinearity in QDs.
The small-sized agglomerates of mercury telluride (HgTe) attract attention as efficient materials for mid-IR applications. Among numerous NPs studied during the last few decades, the exfoliated small-sized structures can be of especial interest due to their applications as efficient emitters of the coherent short-wavelength radiation during high-order harmonics generation. The latter option can be realized during ablation of bismuth telluride (Bi2Te3) NPs contained targets and propagation of laser pulses through the plasmas containing those nanostructures. The interest in the exfoliated Bi2Te3 NPs, apart from the above-mentioned areas, is attributed to the improvement of the characteristics of IR radiation detectors. Bi2Te3 was used as a photosensitive component in the form of epitaxial layers or individual NPs: nanowires or nanoplates. Correspondingly, the use of topological insulators, and in particular bismuth telluride, is actively developing for applications in various areas of electronics. The thin films comprising such NPs are of special interest since they can be incorporated inside the master oscillators to modify the temporal shape of the emitted radiation. All the above-mentioned potential applications of exfoliated Bi2Te3 NPs thin films require the study of their third-order nonlinear optical properties. The use of QDs with a certain set

of localized states causes a strong influence on the formation of accumulative nonlinearities that occur when exposed to nanosecond pulses. Among them are the resonant two-photon excitation and reverse saturable absorption, as well as the nonlinear refraction due to the zone filling effect caused by changes in the population of the dimensional quantization levels and local states in the QD. Such quantum dots include nanocrystals of non-stoichiometric compounds that have a developed system of structural defects involved in the formation of nonlinear refraction. Meanwhile, to the best of our knowledge, there are no studies devoted to the analysis of the low-order nonlinear optical properties of the mercury selenide (HgSe) colloidal QDs. Knowing the nonlinear optical refraction and absorption allows predicting the behavior of the response of these QDs in the case of relatively strong optical fields. This knowledge can be useful during optimization of the potential applications of HgSe QDs, particularly, as the efficient sources of coherent extreme ultraviolet radiation. HgSe colloidal quantum dots allow tailoring their various physical properties by varying the size of the semiconductor core with some additional tuning by modification of the ligand shell. Mercury selenide is also suggested to be used as a topological insulator, with potential for application in spintronics.
Mercury sulfide (HgS) is material useful in various areas of optoelectronics. Different modifications of this material like α-HgS (cinnabar, hexagonal) and β-HgS (meta cinnabar, zinc-blend type, cubic) have been extensively investigated. α-HgS is a wide bandgap semiconductor (E = 2.0 eV), but it converts to the zinc-blende modification (β-HgS) at temperatures above 344 ℃ and becomes a narrow band gap semimetal (E = 0.5 eV). It is a promising material for solar cells and sensors. Bulk mercury sulfide is also suggested to be a topological insulator with potential applications in spintronics. The nanostructures of HgS allow further applications of this material in different fields. Special attention attracts cinnabar nanocrystals that crystallize in the trigonal system with a noncentrosymmetric structure. Meanwhile, HgS colloidal quantum dots remain much less studied semiconductors in comparison to other semiconductor sulfides like PbS, ZnS, or CdS.
Above mentioned materials (QDs and NPs) in form of the thin film (70 − 200 nm) on the 150 μm glass substrate were investigated for nonlinear absorption, refraction and third-order harmonic generation (THG). Schematic view of the experimental arrangement for THG and Z-scan (nonlinear absorption and refraction) are presented on Figure 1, panels (a) and (b) correspondingly.

Fig. 1. Experimental arrangements for the Z-scans (nonlinear absorption and refraction) and THG. (a) Z-scan scheme. PP, probe pulse (150 fs, 500 − 1700 nm); FL, focusing lens; PD1 − PD3, photodiodes; S, sample; TS, translating stage. (b) Experimental setup for third-harmonic generation in thin films. FL: femtosecond laser (150 fs, 100 nJ, λ = 900 - 1700 nm, 500 kHz); QWP: quarter-wave plate; NDF: neutral density filters; L1: focusing lens; S: sample (thin film deposited on the 0.15 mm thick silica glass plate); TS: translating stage; UVF: ultraviolet filter; L2: collecting lens; R: registrar of third harmonic emission (spectrometer USB 2000).

Two tunable lasers were used during these studies. The picosecond laser (EKSPLA PG400 Series Optical Parametric Generator with PL2210 series Picosecond Pump laser) allowed generation of the spectrally tunable pulses. The pulse duration, pulse repetition rate, and spectral range of tuning were 28 ps, 1 kHz, and 400 − 1200 nm, respectively. The femtosecond laser system consisted of ORPHEUS-HP Optical Parametric Amplifier with PHAROS PH2 Femtosecond Pump laser. The generated pulse width was 150 fs with a 500 kHz pulse repetition rate tunable in the range of 400-1700 nm. The temporal and spatial characteristics of laser pulses were maintained approximately same over the whole range of spectral variations of the laser output, which was confirmed by the autocorrelation measurements of pulse width and the beam profiler.
The obtained results have been summarized, presented and published in the three papers [7 - 9], another three papers are submitted and under revision process.

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Abstract ID : 6

Multidisciplinary Laser Spectroscopy

Content

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Multidisciplinary Laser Spectroscopy
Laser spectroscopy is a flourishing research area, which had major impact in science during recent years. On the basic science side, frontlines have been established in fields such as cooling and trapping of atoms and ions resulting in Bose-Einstein condensation, quantum optics and quantum information, as well as high-power laser-matter interaction. In applied laser spectroscopy, the fields of combustion diagnostics, atmospheric remote sensing, agriculture and ecology, as well as biomedicine are prominent. The many aspects of laser spectroscopy are treated, e.g., in [1]. An overview of certain applications of laser spectroscopy is given, with emphasis on the environmental, agricultural/ecological, and biomedical areas is given, based on the experience of the author within these fields at Lund University, and SCNU, Guangzhou. Optical probing of the atmosphere using active remote sensing techniques of the laser-radar type will be discussed [2]. Atmospheric objects of quite varying sizes can be studied. Mercury is the only pollutant in atomic form in the atmosphere, while other pollutants are either molecular or in particle form. Light detection and ranging (Lidar) techniques provide three-dimensional mapping of such constituents. Recently, the techniques have been extended to the ecological field [3]. Monitoring of flying insects and birds is of considerable interest, and several projects have been pursued in collaboration with biologists Fluorescence lidar allows remote monitoring of vegetation and historical building facades. In agricultural applications, e.g., the fertilization levels of crops can be assessed. Drone-based techniques are now also augmenting the possibilities of fluorescence mapping of the environment. Fluorescence spectroscopy has important applications in tissue characterization, using similar methods as for environmental monitoring, but now on a smaller scale. Free gases related to the human body are found, e.g., in the lungs, the middle ear, and the sinus cavities. The gas in scattering media absorption spectroscopy (GASMAS) technique has proved useful in the monitoring of lung function in neonatal children, and shows promising potential in the characterization of otitis and sinusitis [4].
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