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Report

On the summary of the doctoral thesis by Georgi Dobrev

Title: *Laser spectroscopy for coherent manipulation and state-specific probing of atoms and molecules*

General: The doctoralthesis is written in English. It is organized in three parts which describe different experimentsin a context where coherent manipulation of the internal states of quantum systems is essential. Each part starts with its own introductory chapterand concludes with list of references related to the presented study.The summary of the thesis isdistributed on 159 pages and consists of 14 chapters and one Appendix.

The thesis begins with a General introduction section which outlines the structure of the thesis and gives a brief review of the experimental studies performed by Mr. Dobrev,along with a summary of the achieved results.

Part I of the thesis is dedicated on the practical realization of coherent techniques for manipulation of quantum systems by means of a sequence of composite pulses. In the introductory of this part, chapter 1, Mr. Dobrev motivates his choice of ^{40}Ca atom as anappropriatequantum system to validate the concept of composite pulses. In particular exploiting the narrow atomic transition between the singlet $^1S_0$ ground state and the triplet $^3P_1$excited state will strongly reduce the influence of decoherence due to the extended natural lifetime of the excited state. The experimental realization is planned to be executed in an atomic beam. In chapter 2 the author describes the experimental setup, which includes design and construction of the atomic beam apparatus consisting ofa vacuum system anda source ofcalcium atoms. For probing the atoms in the particle beam Mr. Dobrev developed a complete laser system, a diode laser along with the accompanying electronic devices which are needed for its operation. The atomic beam is created from an oven at a temperature of about 600^{o} C, under high-vacuum conditions with a background pressure in the range of 10^{-6} mbar.The oven providesan effusive beam of atoms with an average velocity in the order of 680 m/s. Within this experiment the author tested two different constructions of the atomic source. Due to some technical difficulties, clogging of the oven nozzle occurred in the first developed source making the setup inappropriate for long-term operation. After analyzing the causes of the nozzle clogging Mr. Dobrev provided an improved design of a second atomic source, in which the issue with material crystalizing on the oven nozzle is resolved. The formation of a beam of ^{40}Ca atoms is demonstrated by recording a signal from laser induced fluorescence. The designedtunable single-mode diode laser is employed to excite the $^1S_0$- $^3P_1$intercombination transition in a direction which is orthogonal to the atomic beam propagation. A photomultiplier tube is used to register the weakatomic fluorescence in a phase-sensitive detection scheme. The experimental results from this work are presented in section 2.5. In section 2.6 the author concludes with an extended discussion on the possible causes for the large width of the spectral line which he has obtained and he marks the directions at which the study has to continue.

The current experiment has been performed at the Faculty of Physics in Sofia University “St. Kliment Ohridski”, Sofia, Bulgaria.Although the initial idea for experimental realization of the technique of composite pulses has not been validateditclearly does not undermine the achieved results. For reasons which can not be attributed to him the Mr. Dobrev had to suspend his work on this experiment.

Part II of this thesis presents an experiment in which improvement of the relative frequency stability of a caesium atomic clock is the main concern. The experimental study is performed on the fountain atomic clock CSF2 located at the Physikalisch-Technische Bundesantalt in Braunschweig, Germany.Chapter 3 begins with general motivation for the experiment. In chapter 4Mr. Dobrev provides a detailed information about the electronic structure of the caesium atom. Chapter 5 briefly discusses the techniques for laser cooling and trapping of neutral atoms. In chapter 6 the author provides a detailed description of the construction and the principle of operation of the fountain clock CSF2. Author also introduces the widely used characteristics relative frequency stability and accuracy in the context of fountain clocks and precise absolute frequency measurements. Chapter 7 is devoted to the description of the experiment performed by Mr. Dobrev. In his setup the atomic fountain is operated in the quantum projection noise limited regime where the short term stability (Allan standard deviation) of the frequency measurement is mainly limited by the number of atoms which take part in the Ramsey interrogation cycle. In the CSF2 clock caesium atoms are loaded in an optical molasses arrangementand then the cold ensemble is launched in vertical direction. The atoms traverses twice the same microwave resonator where they experience the Ramsey interaction in a sequence of two $\pi/2$ pulses. In his experiment Mr. Dobrev employs a cold atomic beam in order to provide supplementary source of atoms for loading the optical molasses of the fountain. The atomic beam is created in a modified magneto-optical trap, the so called LVIS – Low-velocity intense source. The LVIS setup is a standard scientific apparatus for producing a beam of slow atoms butit had not been utilized with a great success in atomic clocks.The author's key contribution to this experiment is his proposal of a novel configuration of the laser fields implemented in the original LVIS setup.An additional laser, called "pump" laser,is included along the atomic beam direction. This laser drives the F=4 –> F'=3 transition of the D2line of caesium. Its role is to provide precise control over the velocity of the atoms emerging from the MOT by optically pumping them to a "dark" state, namely the F=3 hyperfine component of the ground state of Cs. In this quantum state the atoms in the beam become insensitive to the accelerating laser which is spatially aligned with the "pump" laser. Another important adjustment of the LVIS-optical molasses system is the change of the orientation of therepumping laser such that it propagates only along the fountain axis. As a result of this the cooled atoms in the optical molasses are concentrated in the vicinity of fountain axis.which improves significantly (factor of two) the number of atoms detected at the end of Ramsey cycle. The employment of the LVIS scheme along with the introduction of the "pump" laser and the changed orientation of the repumping laser in the optical molases gives a factor of 40 riseto the number of atoms which take part in the clock cycle compared to the case of a regular fountain operation. In section 7.3 the described optimization procedures are explained. In section 7.4 Mr. Dobrev presents a qualitative estimations of the parameters mean velocity and flux of the atomic beam. The achieved gain in number of atoms leads to a reduction of the signal-to-noise ratio of the frequency measurement and hence improves the stability of the clock by a factor of six. The measured short-term relative stability of 2.7×10^{-14}at 1s, shown in section 7.5, is a remarkable result, which makes the CSF2 fountain one of the world's most stable primary frequency standards.

Part III of the thesis is devoted to the experimental study of the Zeeman effect in FeH molecule. Species such as FeH and CrH have been identified in spectra of low-temperature stellar objects (sunspots and brown dwarfs) and their potential use as local magnetic probes have been recognized. The complex molecular structure of these compounds makes the exact numerical analysis very difficult and the laboratory study appears as the only opportunity to extract information about transition frequencies, absorption cross-sections and the magnetic properties (Landé factors)of themolecules.Within this study Mr. Dobrev accomplishes several experimental tasks.

The first one includes design and validation of a discharge source which provides sufficient concentration of metal hydride molecules in order to observe absorption spectra both in zero magnetic field as well as in magnetic field up to 0.5T. The work on this task, presented in chapter 11 of the thesis, Mr. Dobrev started in Sofia. He designed a discharge source of a coaxial type which consists of an outer cylindrical cathode, formed from nickel foil with a length of 36 cm, and a central anode part. The NiH molecules are produced by discharge sputtering of cathode material. The source is connected to a primary vacuum system and the discharge is created either in pure H_2 gas or in a mixture of H_2 and Ar gas at a pressure ranging from a few hundred mTorr to 1Torr. In Sofia Mr. Dobrev validated the source by recording differential absorption spectrum of NiH using a tunable diode laser for probing the discharge volume. Later, the source was transferred to Lyon, France where Mr. Dobrev spent 10 months at the University Lyon 1 as an Eiffel scolarschip student. In Lyon he continued his work on improvement of the new discharge source. In the experiments he performed there, Mr. Dobrev used a tunable dye laser to obtain differential absorption spectra of NiH in the (1-0) band between the $B^2\Delta_{5/2}$– $X^2\Delta_{5/2}$ electronic states. The source was also equipped with a magnetic coils which allowed for partially resolved Zeeman spectrum of NiH at a field of 0.1 T to be observed. The purpose of the experiment with NiHis to examine the possibility for using this source for measurements of absolute intensities for transitions of FeH molecule. Although the source provides well defined absorption length and the production rate of molecules is stable over long periods of operation time the sensitivity of the detection scheme restricts the possibility to obtain spectra from the more weakly absorbing molecule FeH. At the end of this chapter, in section 11.4, the author discusses several options to improve the performance of the source by decreasing the cathode length would increase the concentration of molecules in the source volume due to more intense sputtering. He also proposes a scheme consisting of permanent magnets to be used for increasing the strength of the magnetic field inside the source.

Chapter 12 describes an experiment dedicated to the FeH molecule. The molecules are created in a different type of source, which has been used in previous studies of metal hydride molecules performed by the group in Lyon. The hollow-cathode sputtering source produces a stream of molecules in vertical direction and a laser beam, coming out of a tunable Ti:Sapphire laser, probes the molecular jet a few cm below the cathode. An avalanche photodiode is used to register the laser induced fluorescence in phase-sensitive detection scheme. The fluorescence is collected orthogonally to the laser beam and the molecular jet.For investigation of the Zeeman effect a specially designed magnetic circuit is placed below molecular jet. It provides nearly homogenous magnetic field in the laser-molecules interaction region.For calibration of the magnetic fieldseveral atomic transition in Ar with well-known Landé factors are used. The obtained Zeeman spectra of atomic Ar are processed by a curve-fitting routine which calculates the magnitude of the magnetic field with an accuracy of about 1%.Then the same fitting routine is applied to the recorded Zeeman spectra of FeH in order to determine the Landé factors for the rovibrational states of the excited electronic transition which are probed by the laser. The main results of the conducted experiment are the determination of transition frequencies and Landé factors for rotational levels (up to J=10.5)concerning the vibrational levels v=0 and v=1 of the excited electronic state - $F^4\Delta_{7/2}$ of the FeH molecule. The authorworked on routines to simulate and fit complex line profiles in a crowded region of the spectra.This supplied thebest available estimate for Zeeman components of high J lines that could then beincluded in the global fits performed by other members of the Lyon group.

In chapter 13 the author presents laser induced fluorescence spectraof CrH molecule obtained in the sputter source, which was used in the study of FeH radical. By replacing the Fe cathode with one made of chromium Mr. Dobrev successfully obtains a spectrum for several transition lines in the 0 − 0 band of the A $^6\Sigma^+$ –X $^6\Sigma^+$ system of ^{52}CrH and he marks it as a potential subject of future studies of the Zeeman effect in this molecule.

The same sputtering source is used in another experimental setup, described in chapter 14, for measuring absorption coefficients inNiH and NiD molecules. The molecules are formed in similar experimental conditions, in terms of gas pressure and discharge current, as in the case of FeH molecule except that here the cathode material is changed with nickel metal and deuterium is also used as a bufer gas. The discharge source is placed inside a high finesse resonator which allowed for a cavity ring-down spectra to be obtained. The laser source used in this experiment to excite the molecules is a tunable ring dye laser covering the 590-650nm range, and a sophisticated system is developed to control the entire measurement process. The absorption coefficients for the observed spectral lines are derived from the measured ring-down times for each transition. However the absorption coefficient is also a function of the so called "filling factor" which is the ratio of the length of the absorber gas sample to the full length of the cavity. So the accuracy of the estimated absorption coefficients to a great extent is related to the accuracy of determination of the filling factor parameter. As the author discusses this is not an easy task in the particular source especially taking into account that in long-term operation the cathode wears out due to the intensive sputtering, and this alters the production efficiency of the molecules. Nevertheless, the ring-down technique is successfully demonstrated to work with the discharge source and the achieved signal-to-noise ratio of allowed for the spectral lines of the weaker isotopes of nickel ^{62}Ni and ^{64}Ni to be well resolved. The experiment also revealed the possible venues to improve the setup in the future.

In the Appendix of the thesis the author's contributions are summarized followed by lists of publications and conference meetings where the results obtained during his study have been presented.

The amount of work reported within this PhD thesis is enormous. Thediversity of the scientific areas to whichthe three experiments are regarded remind that their successful execution requiresa broad range of skills. G. Dobrev demonstrates expertise in design and construction of different types ofscientific equipment, such as diode lasers, electronic devices, vacuum systems, etc. The author also demonstratesa deep knowledge in atomic and molecular physics. He is well acquainted with experimental techniques applied in various fields such as precise frequency measurements,laser spectroscopy with high resolution, laser cooling and trapping of neutral atoms.I have a very good impression of Mr. Dobrev ever sincethe time when he wasan undergraduate student and I taught him a course in quantum mechanics. Based on the presented thesis I am convinced that during his doctoral study Mr. Georgi Dobrev has molded into an independent experimentalist with creative thinking, which is very important for his personal growth as a future scientist.

My opinion is that the thesis is written clear, the experiments and the results are presented in a logical sequence. Except for a few typographical errors, I did not notice any substantial scientific gaps in the text.

Conclusion: The presented thesis and the number publications of Mr. Georgi Dobrev satisfy the requirements of the national regulations, the internal regulations of the Sofia University "St. Kliment Ohridski" and the recommended criteria by the Faculty of Physics. Regarding the quality of his work I strongly recommend G. Dobrev to be awarded the degree Doctor of Philosophy.

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