Forty years after the first dark resonance experiment: an overview of the COSMA project results

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ABSTRACT

COSMA – Coherent Optics Sensors for Medical Application is an European Marie Curie Project running from 2012 to March 2016, with the participation of 10 teams from Armenia, Bulgaria, India, Israel, Italy, Poland, Russia, UK, USA. The main objective was to focus theoretical and experimental research on biomagnetism phenomena, with the specific aim to develop all-optical sensors dedicated to their detection and suitable for applications in clinical diagnostics. The paper presents some of the most recent results obtained during the exchange visits of the involved scientists, after an introduction about the phenomenon which is the pillar of this kind of research and of many other new fields in laser spectroscopy, atomic physics, and quantum optics: the dark resonance.

Keywords: Coherent Population Trapping, Electromagnetically Induced Transparency, Optical Magnetometry, Nanocell, Coatings, Imaging, All-optical sensors

1. INTRODUCTION

The experiment of Alzetta, Gozzini, Moi, and Orriols[1] was the first experimental observation of the so – called "black line", later indicated as Coherent Population Trapping (CPT), a quantum phenomenon where the destructive interference of the transition amplitudes, when the two laser fields are tuned in resonance with the transitions connecting the hyperfine ground state levels to the excited one, is able to cancel the fluorescence, with a very tight resonance condition. In this way, by the application of both an inhomogeneous magnetic field and multimode, circularly polarized laser radiation to a sodium vapor sealed cell, the authors could directly see by eye one or more thin dark volumes along the dye-laser beam. These dark regions were located in the positions where the frequency distance of two laser modes coincided with the Zeeman shifted distance of atomic levels, to form a lambda system, as shown in fig 1.

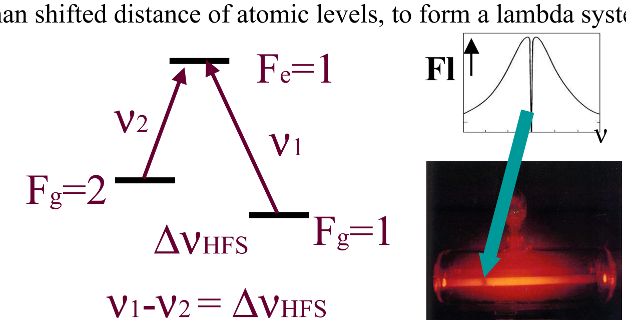

Figure 1. Left: three level scheme for the Sodium excitation; right: picture of the effect and calculation (taken by ref.2) of the transition profile when one of the two laser frequencies is scanned across the resonance.

19th International Conference and School on Quantum Electronics: Laser Physics and Applications, edited by Tanja Dreischuh, Sanka Gateva, Albena Daskalova, Alexandros Serafetinides, Proc. of SPIE Vol. 10226, 102260K · © 2017 SPIE · CCC code: 0277-786X/17/$18 · doi: 10.1117/12.2264896

The interpretation of the ultra – narrow resonant phenomenon, whose reduced bandwidth is related to the very long lifetime of the ground states, was given by Arimondo and Orriols[2], who solved the optical density matrix correlated problem in an analytic way, giving the plot with the typical narrow dip in a much broader structure, obtainable when scanning one of the two laser frequencies across the resonance (upper – right insert of fig.1)

This very clear demonstration of a new effect paved the way for a large variety of applications, still growing in number and importance. We want to briefly give here a few examples of the fantastic consequences this seminal article has left as a legacy.

A particular case of the phenomenon is the use of one of the two lasers as a pump saturating the transition, while using the other one as a probe. This choice has been called Electromagnetically Induced Transparency[3], and recently it has been applied to both a few atoms and even to a single atom stored in a high finesse cavity, merging two innovative fields of quantum optics[4]. One of the most spectacular results of the steep resonance produced by CPT is the possibility to reduce the light velocity to values of the order of tens of meters per second, as demonstrated in [5]. Of course, it is not mandatory to create a three level system playing with the first excited state: in fact, ref. [6] gives a proof of a possible dark state spectroscopy of Rydberg levels, measured as a suppression of the ionization in Neon atoms. The development of more and more advanced atomic clocks, towards outstanding precision limits, able to measure general relativity effect at very small distances, has been favored by using narrow and 100% contrast dark line signal as a reference[7]. It is worth noting that there are also suggestions to implement a CPT – based atomic clock in an undergraduate lab[8], just to underline that both the dark resonances and their applications has become as much "popular" as well as powerful tools. CPT is able to promote four – wave mixing, enhancing the efficiency of the generated signal, as the nonlinear absorption competes with the linear one[9]. The most awarded experiments of the last 20 years, regarding laser cooling and trapping, have got benefit by dark resonances, for example via the Velocity Selective Coherent Population Trapping mechanism[10]. There are also proposals for using EIT spectroscopy in order to detect smaller density molecules in presence of a more populated species[11]. Going to even more complicated physical systems, CPT has been demonstrated in a semiconductor, for a hole spin state in a quantum dot[12]. At completely different energies, EIT has been put in evidence for a Mőssbauer nuclear resonance of Iron, excited by synchrotron radiation in a low finesse cavity where two different samples are obliged to have a superradiant and a subradiant behavior, so to get two different effective excited states, being respectively placed in an antinode and a node of the cavity[13]. Finally, as an introduction to what is commented in the next paragraphs, where an overview of a few of the last year results of the COSMA project will be described, we want to cite the performances of all – optical magnetometers, where the sensitivity of fraction of $fT/\sqrt{Hz}$[14] has allowed for the non cryogenic detection of brain signals with gradiometric configurations.

2. COSMA PROJECT RESULTS

2.1 Study of fundamental processes

One of the possible goal of a fundamental quest of the CPT related processes is the search for new efficient excitation schemes, as well as the comprehension and control of the phenomena involved.

For the formation of high-contrast *N*-type resonance in a Λ-system (ground levels are F_g=2, 3), on the D_1 line of the Rb atoms, two continuous narrow-band diode lasers with λ=795 nm and a 8 mm- long cell filled with the Rb vapor and a buffer gas (20 Torr neon) have been used. A new 4-level system based on an additional (3rd) laser radiation (we call it "preparation" radiation) has been suggested[15]. When the frequency υ_{PREP} is in resonance with rubidium on $5S_{1/2}$, $F_g=2\rightarrow 5P_{3/2}$ transitions (D_2 line, λ=780 nm), via optical pumping process (through $5P_{3/2}$ level) it transfers a large number of Rb atoms from F_g =2 to F_g =3 and creates the inversion condition $N_3(F_g=3) > N_2(F_g=2)$ and if the probe frequency starts from F_g=3, then the amplitude of the "bright" resonance will increase. Meanwhile when frequency υ_{PREP} is in resonance with $5S_{1/2}$, $F_g=3\rightarrow 5P_{3/2}$ transitions it transfers atoms from F_g =3 to F_g =2 and creates the inversion $N_2(F_g=2) > N_3(F_g=3)$. In this case a Raman-type process is switched-on and the initially "bright" resonance is amplified and demonstrates increase in transmission (in the spectrum it looks like a "dark" resonance). Thus, it is possible to control the sign of the resonance.

The good signal/noise ratio of the observed resonance allows us to follow its behavior in an applied magnetic field from several gauss to several hundred gauss. Since narrow-band diode–lasers operating at the wavelengths 780 nm and 795 nm are widely available, the presented system is very robust and convenient for forming controllable narrow-band resonances for practical applications.

While the light – shift problem has been thoroughly investigated in samples confined in uncoated cells, this is not true for the Vector Light Shift (VLS) case in coated vessels. The role of VLS in an optical, synchronously pumped orientation-based Cs magnetometer exploiting a paraffin-coated alkali-metal vapor cell has been discussed in ref. [16]. This kind of magnetometer is based on the resonant response of the atoms to a light modulated at a frequency matching their Larmor-precession frequency. The magnetic field can be extracted by measuring the center frequency of the Lorentzian shaped magnetic resonance. The paper shows that the light shift for a fixed optical power, when the optical pumping is negligible, is independent of the size of the laser beam, with the exception of small systematic contributions. These results are important for modern magnetic sensors that make use of auxiliary fictitious fields and can be extended to other spatially averaged quantities in cells with long coherence times, for example, to the averaging of the scalar and tensor light shifts.

In an alkali dilute gas (for example, Rb and Cs) in presence of a buffer gas, when the exciting laser radiation is tuned to the half-sum frequency of transitions from the two hyperfine sublevels of the ground state, a "bright" resonance can be detected[17]. This "intermediate resonance" is absent in pure vapor cells up to at least 50 mW radiation power, while it is observable in buffered vapor cells already at sub-mW-range power. Due to this fact, together with the existence of a threshold with the laser radiation intensity and the resonant nature of these spectral features, the interpretation is fully or partly attributed to a four-wave mixing process where two mid-resonance laser photons originate two photons on the D2 line hyperfine transitions. This process is strongly assisted by the velocity changing elastic collisions of alkali atoms with the buffer gas ones, which produces an increase of the interaction time.

2.2 Tests of coatings

There are two possible methods in order to both preserve the polarization of the sample as excited by laser light and to eliminate the mixing of the ground state levels due to collision with either the cell walls or with atoms of the same kind: one is to fill the sensor cell with an appropriate buffer gas, the other to coat the cell walls with anti – relaxation coatings. COSMA collaborators have also worked a lot in this second framework.

In order to increase the contrast of the coherent resonance that is one of the main parameters determining the Optical Magnetometer sensitivity an experimental study has been performed and a new theoretical model developed[18]. Here, the formal description of the formation of magneto-optical resonances in alkali-metal atomic vapor is based on a density matrix approach, treated within the formalism of the Liouville equation. The analysis is focused on the case of EIT in spectroscopy cells coated with anti-relaxation polymers. One of the key features, of direct interest for COSMA final goals, is the parametrization of the coating's efficiency: an empirical coefficient ε is in fact introduced, in order to quantify the coating's anti-relaxation properties. The approach allowed the extension of the calculations to many different degrees of depolarization after atom/wall collisions and, therefore, to a broad range of coatings, including uncoated cells.
This provides a formal tool to indirectly evaluate the properties of a coating and thus a quantitative criterion to evaluate its suitability for critical applications, such as ultra-sensitive atomic magnetometry. The investigation reported in Ref. [18], in particular, takes into consideration also different configurations for the EIT formation and different efficiency of the coating characteristics and the atomic energy structure. The model is validated by investigating the EIT with degenerate Zeeman levels in $^{39}K\ D_1$ and Cs D_2 lines, which exhibit respectively an almost negligible and a relevant impact of hyperfine optical pumping. The results are compared to experimental data, exhibiting good agreement and extending the analysis to the $^{39}K\ D_1$ line in the case of degenerate and non-degenerate EIT with amplitude-modulated light.

In this way, more insight in the dynamics of EIT in presence of anti-relaxation coating is provided. In detail, with coatings such as paraffin or polydimethilsiloxane (PDMS), atoms can collide against the cell's walls without any change of their internal state. This implies that atoms, already polarized after interaction with the optical pumping beam, can interact again with the laser beam after bouncing off the wall, without losing their orientation. As a consequence, the angular momentum distribution of the atoms re-entering the laser/atoms interaction volume is a non-equilibrium one: the initial conditions for atoms reentering the laser beam depend on the previous interaction with the laser beam. Whereas in previous works the system's model was solved by the method of iterations, where the number of iterations needed for sufficient convergence is proportional to the number N of collisions of a single atom with walls going without a spin change: ($N = 1/\varepsilon$), thus requiring a large number of iterations for high-quality coatings and practically limiting the applicability ($\varepsilon > 0.1$), in Ref. [18], extension to small values of ε was demonstrated.

A similar approach is pursued also in Ref. [19]: in order to measure the quality of the cell's coating, it is sufficient to measure the ratio between the intensity of the fluorescence at the beginning and at the end of a resonant probing laser pulse, and also the rate of exponential decay of fluorescence intensity during the laser pulse. In this way, a simple method allowing direct study of the dependence of fluorescence spectra on the quality of cell wall coating has been demonstrated. The proposed approach is first demonstrated on the D_1 line of Rb. In the coated cell, it is shown that with irradiation of *Rb* vapor by intensity-modulated laser light, the fluorescence is reduced due to depleting of the excited by the light ground-state level. These coatings could be used for the creation of small sized optical magnetometers and atomic clocks, where by getting higher contrast of the magneto-optical and clock resonances, the accuracy of the magnetic field measurement and time can be improved.

Ref. [20] is devoted to new features observed in the Saturated Absorption spectrum on the D_1 line of K. In an uncoated optical glass cell containing pure K atoms, excitation by circularly polarized pump beam produces an enhancement of the amplitude of the crossover resonances due to the hyperfine transitions starting from the ground state $F_g = 2$. This effect appears to be much more relevant when K atoms are contained in a cell coated with an anti-relaxation film. Here, the crossover resonance in the $F_g = 2$ set of transition is not observed experimentally with linearly polarized pump light, while in case of circular polarization its amplitude is significantly enhanced. The Light Induced Atomic Desorption (LIAD) effect[21] strongly improves the intensities of the Saturated Absorption resonances observed in coated cell.

Narrow magneto-optical resonance on ^{87}Rb D_1 line are investigated in optical cell with paraffin anti – relaxation coating[22]. In two-level degenerated system this resonance is due to the interference between transitions involving the Zeeman sub-levels, created by interaction of resonance linear polarized laser beam with the atoms. The observed signal is detected by sweeping magnetic field **B** around its zero value. In a coated vacuum cell the fluorescence signal has a complex form, because the anti-relaxation coating preserves the created coherence, having different relaxation rates. The ground state coherence is transmitted by the laser field to the upper level; thereby polarization moments with different rank contribute to the fluorescence. The manifestation of the different polarization moments in the observed signal depends on the geometry of the experiment – direction of observation, plane of the laser polarization, polarization of the registered light. The resonances obtained in the fluorescence having different polarization are compared in order to clear up what are the contributions to the fluorescent signal from the polarization moments with different rank. The experiment is performed on the D_1 ^{87}Rb line, $F=2 \rightarrow F'=1$ transition. The magneto-optical resonances, detected in two orthogonal polarizations are measured. Numerical calculations with parameters, close to the experimental ones are performed by using a program, which is based on the irreducible tensor operator formalism. The results of the modeling are in agreement with the measured ones for different experimental conditions.

The observation that, in presence of organic coatings such as paraffin or PDMS, Rb vapor absorption spectra exhibit signatures of a population redistribution produced by optical pumping has been demonstrated in [23]. Deformation of the atomic absorption peaks and shift of their central frequencies have been experimentally correlated to the laser frequency scan rate and direction. These results allow one to investigate the role and the contribution of the coating anti-relaxation properties to the response of the atomic system to optical pumping. In the paper this approach is proposed as a practical, cost- and time-effective method to assess the performance of an organic coating, without hampering the regular experimental activity.

2.3 Miniaturization of sensors – the micro- and nano-cells systems and the related new physics

One of the most intriguing tools of the last 20 years of investigation in the field of high resolution laser spectroscopy is the so – called nanocell, i.e. a special vessel of centimeter size in diameter, but with a thickness limited to lengths ranging from some hundreds of nanometers (the optical radiation wavelength order of magnitude) to micrometric scale.

In this kind of very robust cell (the technical realization allows for temperatures of several hundreds centigrades), atoms show a direct sub – Doppler behavior and CPT/EIT processes can be studied in a new context, that can help a miniaturization of the sensors.

The electromagnetically induced transparency (EIT) phenomenon has been investigated in a Λ-system of the ^{87}Rb D_1 line in an external transverse magnetic field[24]. Two spectroscopic cells having strongly different values of the relaxation rates γ_{rel} have been used: a Rb cell with antirelaxation coating (L ~ 1 cm) and a Rb nanometric − thin cell (nano-cell) with thickness of the atomic vapor column L=795nm. For the EIT in the nano-cell, we have the usual EIT resonances characterized by a reduction in the absorption (i.e. dark resonance (DR)), whereas for the EIT in the Rb cell with an antirelaxation coating, the resonances demonstrate an increase in the absorption (i.e. bright resonances). Such unusual behavior of the EIT resonances (i.e. the reversal of the sign from DR to BR) is likely caused by the influence of alignment process. The influence of alignment strongly depends on the configuration of the coupling and probe frequencies as well as on the configuration of the magnetic field.

Electromagnetically induced transparency (EIT) resonances have been investigated [with the ^{85}Rb D_1 line (795 nm) in strong magnetic fields (up to 2 kG) with three different types of spectroscopic vapor cells: the nano-cell with a thickness along the direction of laser light L ≈795 nm, the micro-cell with L = 30 μm with the addition of a neon buffer gas, and the centimeter-long glass cell[25]. These cells allowed us to observe systematic changes of the EIT spectra when the increasing magnetic field systematically decoupled the total atomic electron and nuclear angular moments (the Paschen-Back/Back-Goudsmit effects). The observations agree well with a theoretical model. The advantages and disadvantages of a particular type of cell are discussed along with the possible practical applications.

The crossover resonances make saturated-absorption spectra very complicated, in particular when an external magnetic field B is applied. Paper [26] is the first experimental demonstration that the use of micrometric-thin cells (in the specific case, the length is around 40 μm) allows for application of saturated absorption in order to quantitatively study both the frequency splitting and the shifts of the Rb atomic transitions in a wide range of external magnetic fields, from 0.2 up to 6 kG (20–600 mT). The authors compare the spectra obtained with the thin cell with those obtained with other techniques. Moreover, they present applications for optical magnetometry with micrometer spatial resolution and a broadly tunable optical frequency lock.

Using a micrometric thickness Cs cell, BAS-UC group recorded sharp EIT and enhanced broad VSOP signals. The observed spectra could be reproduced by a theoretical simulation with a Doppler convolution including all possible orientations of atoms with respect to the laser beam direction[27].

Development of new Magnetometers

UCL designed and developed an alternative experimental approach, based on Magnetic Induction Tomography (MIT), for applications of OAMs in biomedical diagnostics and imaging. The idea is to take advantage of the superior performance of OAMs to measure the small AC field produced by eddy currents excited in the object under evaluation by a primary AC magnetic field (under the operator's control).
The main goal is the imaging of the heart and, in particular, the diagnosis of atrial fibrillation. Nevertheless, other possible applications include detection and imaging of certain classes of tumors, neuropathies and other conditions where the tissues' conductivity is affected.

In order to pursue this goal, an ^{87}Rb OAM operating in self-oscillation regime was designed and tested with the first demonstration of electromagnetic induction imaging with atomic magnetometers[28]. The system was based on amplitude-modulated, synchronous optical pumping of ^{87}Rb atoms contained in a PDMS-coated vapor cell. Although successful, the apparatus required a bulky and expensive phase-locked loop with a PID controller in order to ensure the self-oscillation operation. In view of practical applications in clinical practice and surgery theaters, this could represent a limitation and a source of unreliability.

Therefore, the OAM was completely redesigned in order to reduce the sensor's footprint and cost and to increase its robustness and simplicity: the system's architecture was thus moved to an ^{87}Rb optically-pumped radio-frequency

OAM[29], with several technical improvements in view of operation in 'real-life' situation, in an unshielded environment and at room temperature. As a result of this process, a cost reduction of more than $6000 per sensor unit can be estimated. The new setup is built around a custom-designed cubic vapor cell containing 20 Torr of N_2 as buffer gas. The operating principle is now based on continuous optical pumping by a circular polarized laser beam and coherent driving of the atomic spin population by means of a perpendicular RF field. On the one hand, this allowed further simplification of the system, while maintaining or possibly expanding its wide tunability. On the other hand, it allowed integration of the MIT primary source in the design of the OAM. A linearly polarized probe beam encodes information about the atomic Larmor precession and, hence, about the magnetic fields to be measured in the oscillation of its polarization plane. By performing space-resolved measurements with a balanced polarimeter, one can thus map the local response of an object of interest to excitation by the MIT primary field. The result is a 2D conductivity map which would reveal local anatomy and, more importantly, anomalous distribution of conductivity.
The capability of the UCL's RF OAM to operate both in 'passive' (i.e. as a pure magnetometer) and in 'active' (i.e. as a detector for induction imaging) configurations in an unshielded environment at room temperature was carefully investigated.
Samples with conductivities differing by more than two orders of magnitude were imaged, sub-millimetric features were observed and the penetration of conductive barriers, mimicking reconstruction plates or surgery tools, was demonstrated, all in real time, without any image processing and no background subtraction.

Contextually, a thorough investigation of the possible applications of the system, of its feasibility in actual healthcare environment and its intrinsic advantages was conducted by the UCL unit. Results were reported in a dedicated paper[30], where the immediate application to diagnosis and investigation of cardiac arrhythmias was taken into consideration. In particular, atrial fibrillation, which affects a large fraction of aging population and has a number of severe complications, in some cases fatal, still lacks a dedicated diagnostic tool. Consequently, while its underlying causes are not yet confirmed, its treatment and patients management are also sub-optimal. However, evidence of a correlation with changes in the conductivity of the heart have been found recently. The OAM imaging system under development at UCL would directly map the heart's conductivity, thus providing new insight of the causes of atrial fibrillation and allowing healthcare professionals a dedicated diagnostic tool and more targeted treatments, either drug-based or surgical.
This approach, with to equivalent currently on the market, would be complementary to more conventional investigations such as electrocardiograms and the magneto-cardiogram investigated by other partners of COSMA.

The activity carried out in the last year in the DIISM-UNISI group has been focused on several aspects concerning both applications and fundamental studies and characterization of a Bell-Bloom implementation of a multichannel differential setup magnetometer. The system is based on two gas-buffered Cs cells, where the buffer gas makes the atom motion essentially diffusive and thus different areas of the sample are sensitive only to the respective local field. This feature made possible to analyze separately the interaction in different areas of the probe-beam spot, and to achieve a differential response even with a single cell operation. In conclusion, a dual cell / quadruple channel operation was demonstrated and characterized[31].
Such system enables to get a response to different kinds of field inhomogeneities, detecting selectable higher order derivatives of the variations of a field component.

A theoretical model[32] was developed in order to evaluate the role of the pumping laser on the atomic population/polarization dynamics. In particular, as the modulated parameter of the pumping radiation is the optical frequency (not the amplitude, as in large part of similar setups reported in the literature) it is possible to use the pump radiation to produce both Zeeman pumping and hyperfine pumping, taking advantage from the latter. It was demonstrated that an appropriate choice of the modulation parameters allows for both improving the detected signal and for counteracting relaxation phenomena due to spin-exchange collisions: it is possible to induce the phenomenon known as light-narrowing.

An all-optical ^{133}Cs scalar magnetometer, that operates in nonzero magnetic field, was built at UCB[33]. The system is based on a magnetic resonance driven by an effective oscillating magnetic field that is produced by the AC Stark shift of an intensity-modulated laser beam. The projected shot-noise-limited sensitivity is 1.7 fT/√Hz while the technical noise floor is 40 fT/√Hz. These results reproduce what has been achieved with a coil-driven scalar magnetometer using the same setup. However, the all-optical scheme can be more easily used in arrays and in magnetically sensitive fundamental physics experiments, e.g., searches for a permanent electric dipole moment of the neutron.

The measure of the B-vector is the aim of the last example reported here. If the main interest is not an optimized sensitivity, but a wide field measurement and an experimental simplicity, the two axis optical magnetometer of ref. [34] can be a good solution, especially for geophysical and material science studies. The prototype, based on double scanning of a magnetic field and a nonlinear Hanle effect monitoring, has an accuracy of 5 mG over 1.5 G range, with a huge possible improvement by a faster data acquisition system and the feasible extension to the third B field component.

3. CONCLUSIONS

The work spanned during this last two years of the project with some important realizations of the objectives of COSMA: we developed new collaborations, in particular we strengthened the existing ones, we exchanged and trained young researchers, we had significant exchanges of experienced researchers, we finally extended the scientific level of participating team to detection and analysis of biomagnetism. All the obtained scientific results have to be considered a heritage of the original, apparently simple, setup built 40 years ago in Pisa, as a fruit of the brilliant ideas of A. Gozzini and coworkers.

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