UNIVERSITY OF PÉCS
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Coherent Control of Atomic Quantum States using Frequency-chirped Laser Pulses

PhD Thesis

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Introduction

Understanding the interaction of matter and light was the problem which led to the birth of quantum mechanics a hundred year ago. One of the most important discoveries which burst the frames of classical physics was the discovery of spectral lines of atoms. The first model suitable to explain this observation was by Niels Bohr in 1913: according to his model, only specific orbits of electrons can exist inside an atom, with well-defined energies. When jumping between the orbits, the electron would absorb or emit light corresponding to the energy difference of the orbits.

In 1917, as an extension to Bohr's model, Einstein introduced the concepts of absorption, and spontaneous and stimulated emission, with which he created the foundation for the discovery of the laser.

The appearance of lasers created new directions for the investigations of matter-light interactions by providing an intense, monochromatic and coherent light source. By using laser radiation, better and better coherent control of quantum states of atoms or atomic ensembles can be achieved since it becomes possible to address specific atomic transitions.

By now, laser control of atoms has become an important and rapidly developing field of quantum optics. One of the reasons is that the basic concepts of quantum mechanics can be spectacularly demonstrated by experiments on atom-laser interaction. The significance of such investigations was also highlighted recently by the Nobel-prize.

An important subfield of laser control of atoms is the control of the atoms' valence electrons using laser radiation. The development of this subject is motivated by the possibility of numerous important applications in various fields of modern physics [1–4].

By coherently preparing the atoms, one can modify the optical (refractive and absorptive) properties of the medium composed of said atoms. As a result, several interesting and important nonlinear optical effects may occur or be enhanced [5–10].

The ability to precisely control internal atomic states can be also widely applied in quantum information science. Using atoms as quantum memory devices seems to be especially promising [11–13].

Previous work in the field

There is a large variety of techniques for the coherent control of atomic states. The simplest solution is applying a laser pulse resonant with the atomic transition to be addressed. In this case Rabi-oscillations occur between the coupled states and the proportion of the populations in each state after the interaction depends on the area of the coupling pulse [14]. For example, if this area equals an odd integer multiple of π, complete population inversion can be established. The drawback of this method is that it is very sensitive even to relatively small variations of the pulse area and to the resonance conditions, which may result in additional difficulties for certain experimental setups.

In contrast, coherent control schemes based on adiabatic following, being less sensitive to variations in the experimental parameters, ensure more robust control of atomic quantum states. In these techniques, one of the parameters characteristic to the atom-laser interaction is gradually changed over time, which allows the tuning of the atom's inner state.

One possibility to realize adiabatic following is to change the coupling between the atomic states over time. This is the basic principle of Stimulated Raman Adiabatic Passage (STIRAP) [15,16]. Here the control of a Λ-atom's states is achieved by two time-shifted laser pulses which interact with the atomic transitions in a so-called counter-intuitive order. As a result, the population of one of the ground states is transferred to the other ground state without excitation of the atom, provided that the two-photon resonance condition for the interacting pulses is fulfilled. With extensions to the STIRAP scheme, coherence creation among metastable states of the atom is also possible [17].

Another popular group of the adiabatic control schemes is based on a slowly time-changing detuning between the atomic transition(s) and coupling laser field(s). One possibility for realizing this time-variation is to shift the energy of the atomic levels as is done by a strong non-resonant laser field in the case of Stark-chirped Rapid Adiabatic Passage (SCRAP), making use of the Stark-effect.

An alternative is the temporal modulation of the frequency of the interacting laser pulse(s). The usage of chirped laser pulses for coherent control of atoms has been the main focus of

the group of "Cold plasma and atomic physics" in the Wigner Research Center of Hungarian Academy of Sciences, where I was working as a PhD-student and prepared my dissertation. As direct antecedent to my work, this group made several proposals for solving different control problems, making use of the advantages of chirped pulses.

For example, they proposed an alternative to the STIRAP-scheme by proving that, along with negligible excitation of the atom, complete population-transfer can be achieved between the two ground states of a Λ-atom with a single chirped pulse that couples both atomic transitions [18, 19]. By generalizing this scheme to a tripod-atom they showed that it is also possible to create a coherent superposition among the atomic ground states with a single chirped pulse, again, without significant excitation [20]. The main drawback of the latter method was that the proportion of each state in the created superposition was determined by the properties of the atom (e.g. energy-difference of the atomic levels, strength of the transitions between them).

The advantage of these schemes is that — due to the chirp — they are generally less sensitive to resonance-conditions and thus may be successfully applied in both homogeneously and inhomogeneously broadened media. It is important to avoid the atomic excitation, on one hand, to avoid losing coherence due to spontaneous emission. On the other hand, as was shown in [21] for Λ-atoms, the back-action of the atoms on the interacting laser pulses is small when the atomic excitation is negligible, therefore, this scheme can be also applied for controlling the atoms of a optically thick medium.

By applying more than one laser pulse it becomes possible to reach a wider range of atomic states by varying the parameters of the interacting pulses. For example, when two chirped laser pulses in Raman-resonance interact with a Λ-atom, initially prepared in a coherent superposition of its ground states, the final state of the atom established by these two pulses depends on their constant phase-difference [22, 23]. Making use of this peculiarity, the scheme was proposed to be used for encoding of optical information into quantum states. The drawback of this scheme is that the atom gets excited during the interaction.

Objectives

Motivated by these results, I concentrated in my thesis, on one hand, on upgrading the previous control schemes using FC pulses in order to unify their advantages and to eliminate their drawbacks. That is, one of my objectives was to develop a control scheme using a limited number of chirped pulses which possesses the following characteristics. It is suitable for creating coherent superpositions of the ground state of the atom, which can be adjusted by easily controllable parameters of the interacting laser pulses such as the peak intensities or constant phase difference. In order to avoid decoherence effects due to the finite lifetime of the excited state, I regarded the avoiding of the excitation of the atom as a top priority.

Another goal of mine was to widen the range of possible applicability of chirped pulses by combining them with constant-frequency pulses. I aimed to consider control schemes in Λ-atoms which incorporate instantaneous Raman-resonance and to prove them useful in applications.

In optically thick media, the coherent control processes may be modified by the back-action of the medium on the interacting pulses and other propagation effects. For nonlinear optical applications it is necessary to take these effects into consideration. In my work, I had the aim to discover an interaction scheme using chirped pulses which is applicable for preparing the majority of the atoms in a coherent superposition of their ground states, in an externally adjustable way if possible.

Applied scientific methods

In my thesis, I studied the interaction of atoms and atomic ensembles with a few (2 or 3) frequency-modulated optical laser pulses in a semiclassical approach. From all the effects that the electromagnetic field can possible have on atoms, I was interested in the transitions of the valence electron among the atomic levels. The atoms are modeled as few dimensional quantum-systems. This is possible because the considered laser pulses are all close to resonance with a limited number of atomic transitions and thus transitions are only induced with a non-negligible probability among the corresponding atomic levels. The interacting laser pulses are

strong, hence they can be well described as a classical potential in the Hamiltonian describing the atom-laser interaction.

As the wavelength of optical electromagnetic fields is several orders of magnitude greater than the size of an atom, the atoms can be treated as point-like compared to the interacting light, which allows the application of the dipole-approximation for describing the atom-field interaction. The coupling terms (Rabi-frequencies) between the atomic levels, which represents the interaction of the atoms with the laser pulses, are small compared to the frequency of the lasers and change slowly compared to the timescale defined by the reciprocal of the optical frequency. In order to be consistent with the few-state atom models, the detuning between the frequency of the interacting laser pulses (chirp included) and the frequencies of the atomic transitions driven by them must remain several orders of magnitude smaller than the pulses' frequency during the whole interaction. These features of the interaction described here allows for the application of the rotating wave approximation.

In the majority of the schemes investigated in my dissertation, I assumed that each atomic transition is driven by a separate laser pulse. This assumption can be realized for example by using laser pulses with different polarization for inducing transitions between Zeeman-sublevels.

In the first part of the thesis, when my aim was to create schemes applicable for the coherent preparation of a single atom (or an optically dilute medium), I have described the effect of the laser pulses on the atoms in an interaction picture, by regarding them as external parameters. The frequency-modulation of the pulses was taken into account by time-varing phase of the coupling terms (Rabi-frequencies).

When investigating the interaction between a tripod-atom and three chirped pulses (theses 1. and 2.), I described the atomic dynamics using the master-equation, taking into account the relaxation processes (both longitudinal and transverse) that occur inside the system, but neglecting the decay towards the environment. The equations were numerically solved with parameters plausible in experimental situations. The effect of certain factors (spontaneous emission, dephasing, Doppler-broadening, parameters of laser pulses) on the proposed control schemes was investigated. In the case of Doppler-broadening I averaged the results for different values of detuning by weighting them with a probability-function obtained from the Maxwell-

Boltzmann distribution.

In the case of the interaction of a Λ-atom with a combination of a chirped and a constant-frequency pulse (theses 3. and 4.), I made the assumption that the interaction time is significantly shorter than the lifetime of the excited state and I calculated the atomic dynamics by numerically solving the Schródinger-equation. The effect of the variation in the interaction parameters was again investigated by numerical simulations for a wide range of parameter values.

In thesis 4., I investigated the interaction of an atom having two ground states with a combination of a chirped and a constant-frequency pulse, both far detuned from the possible atomic transitions, through the example of the $|5S_{1/2}, F = 1\rangle \rightarrow |5P_{3/2}\rangle$ transition of ^{87}Rb. I took into account all the possible atomic transitions allowed by the selection rules, which resulted in a 7-level atom model.

When describing the interaction of a pair of Raman-resonant chirped pulses with an optically dense medium (thesis 5.), it was necessary to take into account the back-action of the medium on the electromagnetic fields. Therefore, the model treats the Rabi-frequencies as dynamical variables (as opposed to external parameters) in this model, of which the dynamics is given by the Maxwell-equations in the frame of slowly varying envelope approximation. The effect of the medium's atoms on the laser pulses is described by the macroscopic polarization. This quantity is given in each spatial location by the average of the dipole moments of the atoms which are in the infinitesimal proximity of the said location. The slowly varying envelope approximation could be applied as the envelope of the laser pulses was slowly changing on the scales defined by the frequency and wavelength of the laser pulses.

The majority of the interaction schemes proposed in this dissertation were based on the gradual (adiabatic) change of the interacting laser pulses' frequency. Therefore, the dynamics of the atomic states could be described by using the adiabatic approximation. This approximation gives information about the change of the atomic states over time by analyzing the eigenvalues and eigenvectors of the interaction Hamiltonian of the atom-laser system. In addition to the numerical methods described above, I also applied the 'adiabatic following method' for the majority of the schemes in order to describe the underlying physical mechanisms behind the

numerical results.

Theses

1. I have proposed a new interaction scheme for creating coherent superposition among the metastable ('ground') states of an atom having tripod-structure as working levels ('tripod-atom'), along with negligible excitation. The method is based on adiabatic control of the atomic states by three laser pulses with the same (linear) modulation ('chirp') in their carrier frequencies, each of them separately coupling one dipole allowed transition of the atom. I have shown by analyzing the time evolution of the adiabatic states that if two pulses are in Raman-resonance with the corresponding atomic transitions and the third one is Raman-detuned, two different population-redistribution mechanism may be achieved by changing the sign of the Raman-detuning, without significant population appearing in the excited state during the interaction.

   By numerically solving the master-equation of the system, I have demonstrated that the scheme can be used even if the lifetime of the excited state is one tenth of the interaction time. I have found that coherence can also be created between the ground state in the presence of stronger spontaneous decay from the excited state, with a factor of π difference in phase compared to the non-decaying case.

   I have demonstrated that the presented scheme may be applicable in an (optically dilute) atomic gas at room temperature in the case of fast enough frequency modulation and copropagating pulses, since the chirp is capable to compensate for the Doppler-effect. [I, II]

2. I have proposed a method for robust writing and storage of optical phase information in the populations of the ground states of a tripod atom using three frequency-chirped laser pulses, two in Raman resonance and the third Raman-detuned from the corresponding atomic transition (the same arrangement as in thesis 1). The scheme is based on the fact that after preparing the atom in a coherent superposition of the two ground states coupled by two pulses in Raman resonance, the population of the third ground state

established by the interacting pulses is a cosine-function of the phase difference of the pulses in Raman resonance. Since the information is mapped to the populations of the atomic ground states, the proposed method provides much longer storage times compared to the schemes based on collective atomic spin coherences, being only restricted by the lifetime of the ground states.

I have shown that the presence of weak longitudinal and transverse relaxation affects the contrast of the mapping process: 50% of contrast can be achieved if the duration of the interaction is smaller than a tenth of the coherence lifetime and 10 times the excited state lifetime, respectively [III].

3. I have demonstrated that a pair of laser pulses, one with constant and the other with monotonously modulated carrier frequency, establishes a maximum coherence between a ground and the excited state of a λ-atom. The initially unpopulated ground state is resonantly coupled by a constant-frequency laser pulse to the excited state, forming an Autler-Townes doublet, while the other pulse having a frequency sweeping through one-photon and two photon resonance adiabatically transfers the population from the ground state to that member of the doublet with which it first become resonant. I have shown that the mechanism, provided the resonance condition of the constant-frequency pulse is fulfilled, is extremely robust to the parameters of the laser pulses such as the Rabi-frequencies or the chirp rate [IV].

4. I have proposed a novel scheme for creating coherent superposition between two magnetic sublevels of the $F = 1$ hyperfine ground state of ^{87}Rb atoms. It is based on the interaction of the atom with a frequency-modulated (chirped) laser pulse having σ^+ and a pulse of a constant carrier-frequency having σ^- circular polarization, both far detuned from one-photon resonance but with the chirped pulse sweeping through two-photon resonance. Taking into account all the possible atomic transitions allowed by the selection rules, I have shown numerically that the scheme yields a maximum possible degree of coherence of superposed states with negligible atomic excitation, which eliminates decoherence by spontaneous emission [V].

5. I have demonstrated a matching effect for a pair of frequency-modulated (chirped) Raman-resonant laser pulses which simultaneously propagate in an optically thick medium of lambda-structured atoms. That is, the laser pulses become distorted by the interaction with the medium in such a way that it no longer causes excitation in the atoms, reducing the back-action of the atoms on the pulses and allowing a quasi-lossless propagation. By introducing a coordinate transformation, I separated a symmetric and an asymmetric mode of the lasers, with the latter being 0 at the boundary of the medium. I have shown that due to the excitation of the atoms at the boundary, the asymmetric mode appears, changing the population transfer mechanism in such a way that the majority of the population is transferred to the same coherent superposition of the atomic ground states throughout the medium. I have also shown that the composition of this coherent superposition can be controlled by the peak amplitudes of the these laser pulses at the boundary of the medium [VI,VII].

Conclusion

In my PhD-thesis, I investigated adiabatic control schemes — suitable for preparation of atoms or atomic ensembles in specific internal states — based on using frequency-modulated laser pulses. I have shown that the proposed schemes are suitable for robustly creating coherent superpositions between atomic quantum states in a way that it is possible to adjust the proportion of each state in the superposition by external parameters (the proportion of the peak amplitudes, and the difference of the complex phases of the interacting laser pulses). Since these parameters are relatively easy to adjust in experiments, these schemes may easily find applications in nonlinear optics or quantum information science. I also proposed a possible application of the scheme for optical information writing.

By analyzing the propagation of a Raman-resonant chirped pulse pair in a medium consisting of Λ-atoms I showed that control of the state of the atoms within the medium can be achieved with a good efficiency. I have found that the chirped pulse pair can propagate in a quasi-lossless way in the medium due to a self-organizing (matching) mechanism which is different from the

effect known for the propagation of constant-frequency pulses.

The physical system which served as a base of my investigations was a cold atomic cloud of alkali atoms. More specifically, I modeled transitions of the atoms' valence electron between their metastable and first excited states. In future work I plan to investigate the applicability of these schemes to other physical systems. One possible aim may be to create Rydberg-excitations in atoms by applying chirped pulses. Another possible direction is to find applications of the proposed schemes in solid-state systems, especially for the control of the states of artificial atoms created in quantum dots.

Publications

Publications related to the theses:

[I] N. Sandor, J. S. Bakos, Zs. Sörlei, and G. P. Djotyan,Creation of coherent superposition states in inhomogeneously broadened media with relaxation. J. Opt. Soc. Am. B, 28:2785-2796, (2011).

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