5.1 Allgemeine Angaben zum beendeten Teilprojekt C8

5.1.1 Titel:

Control of light by ultracold matter

5.1.2 Fachgebiete und Arbeitsrichtung:

Theoretical atomic and molecular physics, optimal control theory, light-matter interaction

5.1.3 Leiter:

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5.2 Bericht über die Entwicklung des Teilprojekts

5.2.1 Bericht

Coherent control was constructed to steer a quantum system from an initial state to a final objective via an external field. The idea is to control the interference pathway governing the dynamics from the initial to the final state. For pure initial and final states the method can be termed state-to-state coherent control. A generalization is steering simultaneously a set of initial pure states to a set of final states, i.e. controlling a unitary transformation. Such an application sets the foundation for a quantum gate operation. Two basic questions address applicability of coherent control. The first: For a preset initial and target state does a control field exist? This is the problem of controllability. The second: How to find the optimal field

that carries out this task? This is the problem of Optimal Control Theory. Experimentally there has been remarkable success in constructing devices able to generate arbitrary control fields. Nevertheless experimentally complete controllability is hard to get even in small quantum systems.

The goal of this project was to study coherent control of the light-matter interaction to achieve (A) light-shaping in the UV and (B) ultrafast optical detection for ultracold molecules and molecular Bose-Einstein condensates. This was to be explored for alkali atoms and dimers in collaboration with project A1 (Wöste/Lindinger). Non-resonant multi-photon transitions leading to harmonic generation of order three to nine represent the key ingredient. Their theoretical description as well as the optimal control theory of time-dependent targets was studied in collaboration with projects C3 (May).

Coherent control has provided the framework for both projects. Specifically, the atomic or molecular transition dipole or the excited state population need to be controlled in a time-dependent fashion. Such a time-dependent target functional results in an inhomogeneous Schrödinger equation that needs to be solved as part of the optimization algorithm [NTK09]. Moreover, no monotonically convergent algorithm for a non-positive semi-definite operator such as the transition dipole was available. This has lead us to revisit Krotov's method for optimal control and derive generally applicable, stable, efficient and monotonically convergent optimization algorithms for quantum control problems [RNK10].

Coherent control ideas can be extended to shaping matter wave with time-dependent potentials. Our goal was to supply design ideas for future experiments on atom chips [NJF07]. Assuming a specific setup on an atom chip, we explored the ability to focus, accelerate, reflect, and stop an atomic wave packet. The controls considered were varying electric fields from an array of electrodes. We also utilized this method to initiate coherent splitting or an arbitrary wave form, cf. Figure 1. Special emphasis was devoted to the robustness of the control schemes. We begin with the wave packet of a single atom and extend this to a Bose-Einstein condensate in the Gross-Pitaevskii picture. In analogy to laser pulse shaping with its wide variety of applications, we expect this work to form the base for more complex time-dependent potentials, eventually leading to matter-wave pulse shaping with numerous applications [NJF07]. Controlling directly matter waves is a new direction for coherent control. It relies on recent progress in cooling and the possibility of generating degenerate quantum gases. At this point in time we are on the verge of a new era of Bose-Einstein condensate of molecules. This will open new horizons of shaping matter waves consisting of molecules.

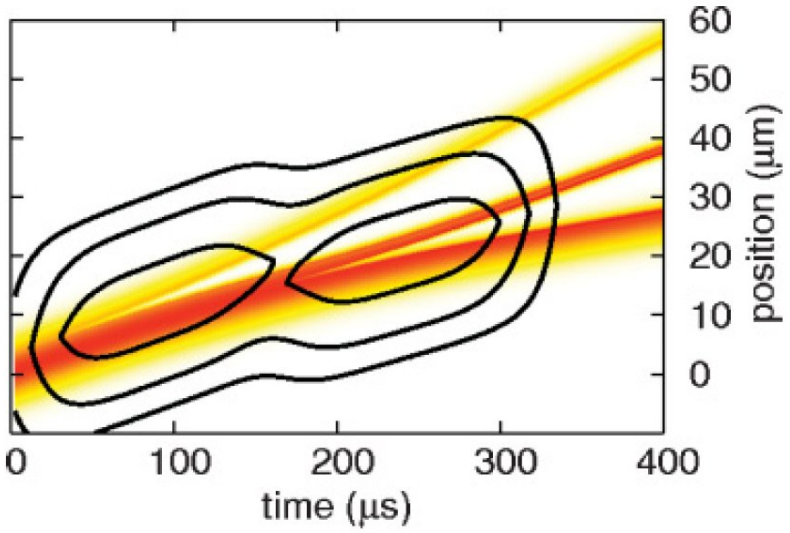

Figure 1: Asymmetric splitting of the wave packet into three parts. Taken from Ref. [1].

The scaling of a quantum control task with increasing Hilbert space size was studied [KKo10]. The double well Bose Hubbard model was employed as the model. This task is related to the issue of matter wave coherent control. The model can be realized as a Bose-Einstein condensate in a double well potential. For initial and target states that are classified as generalized coherent states (GCS) a scalable control field was found. For such problems, a pilot field generated for a small system is adequate also for larger systems. No scaling properties are found for superpositions of GCS or cat states. This study is part of a theme to discover the limits of controllable systems. The scaling of the search effort to obtain a control field with the system size was found to be divided into cases where the initial and target state were GCS or generated from GCS by symmetry and superpositions of GCS. In the first category a pilot field based on maximum G-purity GCS scaled up with the system size so that the control fields could be obtained for very large systems. The second category showed no scalable features of the field. This means that in this su(N) system the task of generating a quantum compiler, translating an arbitrary unitary transformation to a control field will scale unfavorably with system size. The present finding is consistent with the analysis that noise in the control field for large system size will transform any superposition of GCS states to a mixture thus further degrading the controllability.

Optimal control theory for time-dependent targets such as a specific time-dependence of the dipole operator to generate control of light emission, requires the ability to solve inhomogeneous Schrödinger equations. A stable, accurate and efficient propagation scheme for time-dependent inhomogeneous Schrödinger equations was developed in Ref. [NTK09].

Such equations occur also in reactive scattering or in optimal control theory with an intermediate-time cost that depends explicitly on the state of the system. Functionals of this type are obtained for example when prescribing a certain 'trajectory', optimizing a time-dependent expectation value or keeping the system dynamics in a certain subspace of the full Hilbert space. A formal solution based on a polynomial expansion of the inhomogeneous term was presented in Ref. [NTK09]. It is subjected to an approximation in terms of Chebychev polynomials. Different variants for the inhomogeneous propagator are demonstrated and applied to two examples from optimal control theory. The convergence behavior and the numerical efficiency were analyzed.

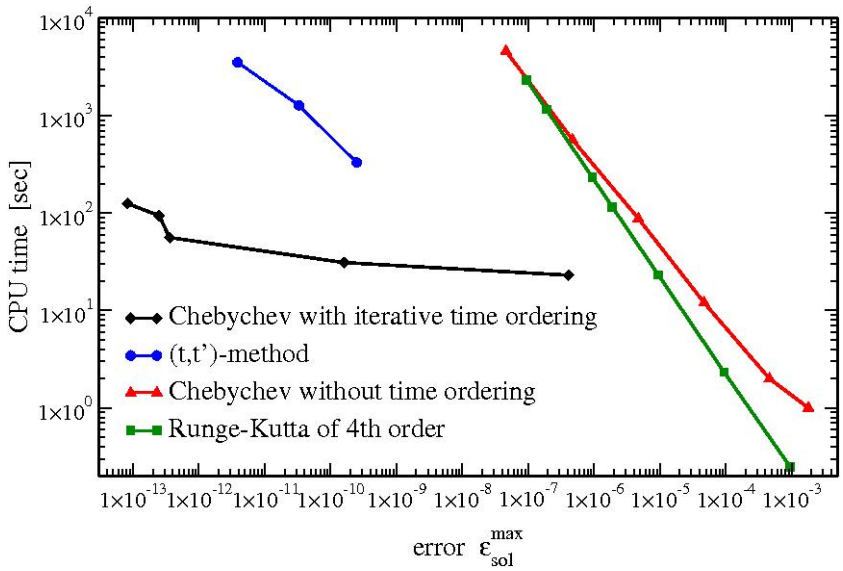

Figure 2: Comparison of propagation methods for strongly time-dependent Hamiltonian. The Chebychev propagator with iterativve time-ordering outperforms all other methods in terms of numerical efficiency if high accuracy is required. Taken from Ref. [5].

The propagation scheme of Ref. [NTK09] can be utilized to solve the Schrödinger equation with explicitly time-dependent Hamiltonian [NTK10]. Time ordering is achieved iteratively. The explicit time dependence of the time-dependent Schrödinger equation is rewritten as an inhomogeneous term. At each step of the iteration, the resulting inhomogeneous Schrödinger equation is solved with the Chebychev propagation scheme presented in Ref. [NTK09]. The iteratively time-ordering Chebychev propagator was shown to be robust, efficient, and accurate and compares very favorably with all other available propagation schemes, cf. Figure 2.

The most prominent example where strong time-dependencies occur and require to take time ordering into account is optimal control. When applying the propagators developed in [NTK09, NTK10] to optimal control, an additional difficulty is posed by the fact that the

time-dependence which is due to the optimized field is not known in advance, but is determined iteratively. Combination of the newly developed propagators [NTK09, NTK10] with optimal control theory therefore requires an additional iterative loop, similar to the one employed in Ref. [NTK10] when rewriting the time-dependence as an inhomogeneity. This is the subject of a currently on-going study.

Generally, quantum control problems can be classified according to the optimization functionals, equations of motion and dependency of the Hamiltonian on the control. Using a second order construction, we have derived a class of monotonically convergent optimization algorithms [RNK10]. We have shown that for most quantum control problems, the second order contribution can be straightforwardly estimated since optimization is performed over compact sets of candidate states. For each problem class of quantum control problems, we have outlined the resulting monotonically convergent algorithm. While a second order construction is necessary to ensure monotonic convergence in general, for the ’standard’ quantum control problem of a convex final-time functional, linear equations of motion and linear dependency of the Hamiltonian on the field, both first and second order algorithms converge monotonically. We have compared convergence behaviour and performance of first and second order algorithms for two generic optimization examples [RNK10]. This work paves the way to study optimal control of a very broad range of problems including light emission and quantum state detection.

Photoassociation with short laser pulses has been proposed as a technique to create ultracold ground state molecules. In the general effort to understand photoassociation processes taking place in ultracold gases we investigated a new possible scheme. Bound-state molecules can be photoassociated directly from ultracold free-atom pairs by excitation to a purely repulsive electronic state [KKo08]. The process is explained on the basis of quantum unitarity: the initially free-scattering state is transformed by an impulsive light pulse to a deformed superposition which contains bound-state components. For pulse durations which are short compared to the ultracold dynamics, the maximal rate of photoassociation was found to be determined by the initial stationary distribution of scattering states of the atom pairs. The process was simulated for an ultracold gas of ^{87}Rb. Transform-limited pulses maximize the photoassociation, yielding approximately one bound molecule per pulse [KKo08]. Coherent control calculated by a local control scheme can increase the photoassociation yield by two orders of magnitude.

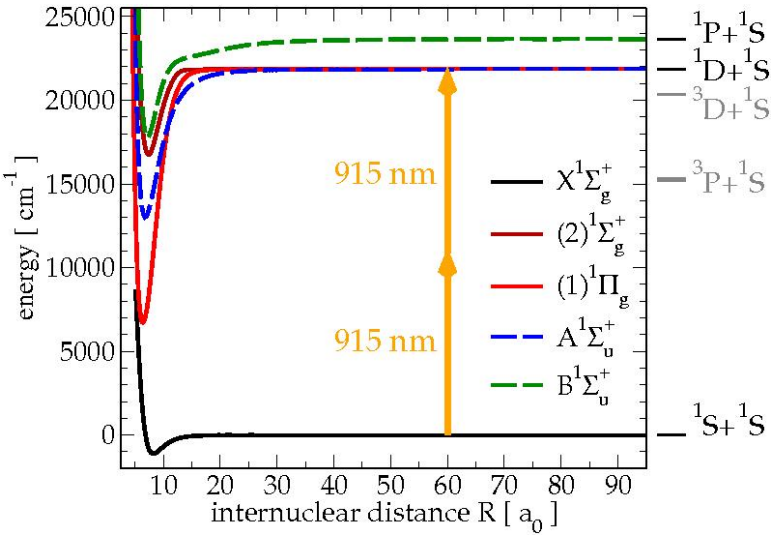

Figure 3: Two-photon photoassociation scheme for broadband excitation. An effective model to describe the two-photon transitions is used. Taken from Ref. [4].

In photoassociation, a broad-band excitation seems the natural choice to drive the series of excitation and deexcitation steps required to form a molecule in its vibronic ground state from two scattering atoms. First attempts at femtosecond photoassociation were, however, hampered by the requirement to eliminate the atomic excitation leading to trap depletion. On the other hand, molecular levels very close to the atomic transition are to be excited. The broad bandwidth of a femtosecond laser then appears to be rather an obstacle. To overcome the ostensible conflict of driving a narrow transition by a broad-band laser, we have suggested a two-photon photoassociation scheme [KNK09]. In the weak-field regime, a spectral phase pattern can be employed to eliminate the atomic line. When the excitation is carried out by more than one photon, different pathways in the field can be interfered constructively or destructively. In the strongfield regime, a temporal phase can be applied to control dynamic Stark shifts. The atomic transition is suppressed by choosing a phase which keeps the levels out of resonance. We derive analytical solutions for atomic two-photon dark states in both the weak-field and strong-field regime. Two-photon excitation may thus pave the way toward coherent control of photoassociation. Ultimately, the success of such a scheme will depend on the details of the excited electronic states and transition dipole moments. We have explored the possibility of two-photon femtosecond photoassociation for alkali and alkaline-earth metal dimers and presented a detailed study for the example of calcium, cf. Figure 3 [KNK09].

We are currently engaged with a similar system of photoassociation of magnesium. We are simulating an experiment carried out by the group of Zohar Amitay in the Technion where a two-photon photoassociation is performed probed by a third probe photon which leads to light

emission. The theoretical work is carried out by Sai Amaran. This system has the potential to realize coherent control with the target of shaping the emitted light.

Photoassociation creates molecules in their electronic ground state which are vibrationally highly excited. In order to utilize the molecules in potential applications, they need to be transferred to their vibrational ground state. The transfer of weakly bound KRb molecules from levels just below the dissociation threshold into the vibrational ground state with shaped laser pulses was studied in Ref. [NKo10]. Optimal control theory was employed to calculate the pulses carrying out the vibrational stabilization. In principle, optimal control theory works independently of the underlying molecular structure. However, the specific outcome of the optimization will of course depend on the molecular model. In Ref. [NKo10] we have studied the influence of the molecular structure on the optimization problem. We have successively increased the complexity of modelling the molecular structure in order to study the effects of the long-range behaviour of the excited state potential, resonant spin-orbit coupling and singlet-triplet mixing. The optimal dynamics was found to be surprisingly little influenced by the specifics of the molecular structure, and the largest effects could be attributed to the size of the underlying Hilbert space.

Summary and outlook

The formation of molecules from ultracold atoms using laser light presents itself as an ideal candidate for coherent control: The process is complex and poses many challenges. A central theme in photoassociation is the interplay between the time scale of the light induced process and the timescale of the free evolution dynamics [KKo08, KKo10]. We can classify the sudden or impulsive region where the pulse is shorter than the free evolution and the opposite long time limit where the light induces transitions on resonance between well defined eigenstates. We developed novel numerical schemes to tackle these problems [NJF07, NTK09, NTK10, RNK10]. These developments are a prerequisite for any further theoretical investigation. In this research we devoted significant effort to the many body aspect of control. This aspect is part of our original intention of controlling emission [KNK09]. We studied the control of matter waves in a Bose Einstein condensate context [NJF07, KKo10]. We find that not all targets are controllable. In this subproject we have just opened the door. Significant research is still required to address the many body aspect of coherent control.

5.2.2 Publications resulting from the project

[NJF07] Mathias Nest, Y. Japha, R. Folman and Ronnie Kosloff
Dynamic matter-wave pulse shaping
Phys. Rev. A. 81, 023632 (2007)

[KKo08] Shimshon Kallush, and Ronnie Kosloff
Unitary photoassociation: One-step production of groundstate bound molecules
Phys. Rev. A 77, 023421 (2008)

[NTK09] Mamadou Ndong, Hillel Tal-Ezer, Ronnie Kosloff, and Christiane P. Koch.
A Chebychev propagator for inhomogeneous Schrödinger equations.
J. Chem. Phys. 130, 124108 (2009), arXiv:0812.4428

[KNK09] Christiane P. Koch, Mamadou Ndong, and Ronnie Kosloff.
Coherent control of two-photon femtosecond photoassociation
Faraday Discussions 142, 389 (2009), arXiv:0810.5738

[NTK10] Mamadou Ndong, Hillel Tal-Ezer, Ronnie Kosloff, and Christiane P. Koch.
A Chebychev propagator for time-dependent Hamiltonians based on iterative time ordering
J. Chem. Phys. 132, 064105 (2010), arXiv:0912.3363

[NKo10] Mamadou Ndong, and Christiane P. Koch.
Vibrational stabilization of ultracold KRb molecules. A comparative study
submitted to Phys. Rev. A (July 2010), arXiv:1007.1898

[RNK10] Daniel Reich, Mamadou Ndong, and Christiane P. Koch.
Monotonically convergent optimization in quantum control using Krotov's method
submitted to J. Chem. Phys. (2010)

[KKo10] Shimshon Kallush, and Ronnie Kosloff
The scaling of quantum controllable problems
submitted, New. J. of Phys. (2010)

5.3 Bewilligte Mittel für die laufende Förderperiode

Das Teilprojekt wurde im Sonderforschungsbereich von 07/2007 bis 06/2010 gefördert.

Haushalts-jahr	Personalmittel	Sachmittel	Investitionsmittel	Gesamt
2007/2	40.2	0	0	40.2
2008	80.4	11.9	0	92.3
2009	80.4	0	0	80.4
2010/1	40.2	0	0	40.2
Summe	241.2	11.9	0	253.1

(Alle Angaben in Tausend EUR)

5.3.1 Personal im Teilprojekt

	Name, akad. Grad, Dienststellung	engeres Fach des Mitarbeiters	Institut der Hochschule oder der außeruniv. Einrichtung	im SFB tätig von (Monat / Jahr) bis (Monat / Jahr)	Entgelt-gruppe
Grundausstattung					
wissenschaftl. Personal (einschl. Hilfskräfte)	1. Koch, Christiane, Dr.	Theoretical Physics	Inst. f. Theor. Physik, FUB	07/2007 – 06/2010	
	2. Kosloff, Ronnie, Prof.	Theoretical Chemistry	Hebrew Uni Jerusalem	07/2007 – 06/2010	
Ergänzungsausstattung					
wissenschaftl. Personal (einschl. Hilfskräfte)	3. Ndong, Mamadou, Dr.	Theoretical Physics	Inst. f. Theor. Physik, FUB	12/2007 – 06/2010	BAT IIa 1/1
	4. Kallus, Shimshon, Dr.	Theoretical Chemistry	Hebrew Uni Jerusalem	07/2007 – 06/2010	BAT IIa 0,35
	5. Amaran, Saieswari, Dr.	Theoretical Chemistry	Hebrew Uni Jerusalem	07/2007 – 06/2010	BAT IIa 0,35