Project plan for Johannes' bachelor thesis: the role of excited atomic states in multiphoton ionization

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1 Project description

The goal of this project is to investigate, within the strong-field approximation (SFA), how the polarization of a hydrogen atom by the electric field of an intense, few-cycle laser pulse, the Stark shift induced by the pulse, and the transitions to excited states affect the rate of multiphoton ionization. In the SFA, the Coulomb interaction between a free electron and its parent ion is neglected. Most importantly in the context of the project, SFA provides a rigorous way to calculate ionization rates, which is not widely known, but it is quite straightforward. Manoram has developed a Python code that performs the calculation under the assumption that the laser field has no effect on the atom until the moment of ionization, i.e., excited atomic states are neglected. In this approximation, the free-electron wavefunction after interaction with the laser pulse is given by the following expression, written in atomic units using LaTeX notation:

$$\lim_{t\to\infty} |\Psi(t)\rangle = -i \int d^3p\, |\mathbf{p}\rangle \int_{-\infty}^{\infty} dt'\, e^{-\frac{i}{2}\int_{t'}^{\infty}[\mathbf{p}+\mathbf{A}(t')]^2\,dt'} e^{iI_\mathrm{p}t'} \langle \mathbf{p}+\mathbf{A}(t')|\hat{\mathbf{d}}\cdot\mathbf{E}(t')|\Psi_0\rangle,$$

where $\mathbf{p}$ is the momentum, $\mathbf{E}(t) = -A'(t)$ is the electric field of the pulse, $\mathbf{A}(t)$ is the vector potential, I_p is the ionization potential, $\hat{\mathbf{d}}$ is the dipole operator, and $|\Psi_0\rangle$ is the wave function that describes the atomic ground state. The key idea of the project is to replace $e^{iI_\mathrm{p}t'}|\Psi_0\rangle$ by a time-dependent superposition of several atomic states $\sum_n c_n(t)\exp\{-iE_nt\}|\Psi_n\rangle$, where the probability interaction-representation amplitudes $c_n(t)$ will be obtained by numerically solving the time-dependent Schrödinger equation (TDSE). An implicit assumption here is that ionization does not significantly change these probability amplitudes, which restricts the analysis to small ionization probabilities. For the numerical solution of the TDSE, we will use one of the existing open-source codes (we have a strong preference for tRecX: https://gitlab.physik.uni-muenchen.de/AG-Scrinzi/tRecX).

As a result of this project, we would like to learn how accounting for the effect of the laser field on a neutral atom changes the ionization rates for different laser pulses. In particular, we hope to learn what matters most: the Stark shift or the distortion of the ground-state wavefunction by the laser field. With some luck, we may even find an analytical approximation that describes such corrections to ionization rates.

2 Project Breakdown and Planning

2.1 Work Package 1: Extraction of Time-Dependent Probability Amplitudes from tRecX

Objective: To develop a robust procedure for extracting the time-dependent probability amplitudes of the ground state and all relevant excited states from the tRecX output files for use in the SFA calculations.

Research Questions:

- How to locate and load the data containing the time-dependent probability amplitudes for different atomic states within the output files of tRecX?
- How to efficiently store and organize the extracted time-dependent probability amplitudes for use in the modified SFA Python code?

Methodology:

1. Store the extracted time-dependent probability amplitudes in a structured format that can be easily accessed by the modified SFA code.
    - Make sure that the probability amplitudes are stored in the interaction representation.
    - Store the energies of the states together with the probability amplitudes to facilitate a possible future generalization of the code to multielectron atoms.

2.2 Work Package 2: Modification of the SFA Python Code

Objective: To modify the existing SFA Python code to incorporate the time-dependent probability amplitudes of the atomic states obtained from tRecX, allowing for a superposition of initial states.

Research Questions:

- How to adapt the existing SFA integral formula to account for a time-dependent initial state that is a superposition of the ground state and relevant excited states?
- How to calculate the dipole matrix elements $\langle \mathbf{p} + \mathbf{A}(t') | \hat{\mathbf{d}} \cdot \mathbf{E}(t') | \Psi_n \rangle$ not only for the ground state ($n = 0$) but also for the relevant excited states ($n > 0$) of hydrogen?
- How to make the necessary changes to the Python code that calculates SFA ionization rates and what tests need to be implemented?
    - Does the integration over angles have to be done before or after calculating the sum over atomic states?

Methodology:

1. Obtain analytical expressions for the dipole matrix elements responsible for the transitions from the eigenstates of the hydrogen atom to plane waves.
2. Analyze the existing SFA Python code.
3. Modify the code to load the time-dependent probability amplitudes for the ground state and the relevant excited states extracted in Work Package 1. Initialize interpolation objects that provide probability amplitudes at arbitrary times.
4. Modify the code to include a sum over the relevant initial states, where each state's contribution is weighted by its time-dependent probability amplitude obtained from tRecX.
5. Adapt the calculation of the ionization rate to account for this superposition of initial states.
6. Test the modified code.

Expected Outcomes:

- A modified and tested Python code capable of calculating ionization rates within the SFA framework, taking into account the time-dependent populations of the ground and relevant excited states of hydrogen obtained from tRecX.
- The new code must provide an easy way to neglect the effect of the laser field on the neutral atom, in which case the data for the time-dependent probability amplitudes will not be loaded, and the results will be exactly equivalent to those produced by the original code.
- The new code must be decoupled from tRecX to make it easy to replace tRecX with other code in the future.

2.3 Work Package 3: Calculation of Ionization Rates and Analysis

Objective: To utilize the modified Python code to calculate ionization rates for a range of laser pulse parameters and analyze the impact of including excited atomic states on the ionization dynamics.

Research Questions:

- How do the calculated SFA ionization rates for hydrogen change when the time-dependent populations of excited states (obtained from tRecX) are incorporated into the initial state?

- Which excited states have the most significant influence on the ionization rates?
- Are there any discernible trends or patterns in the corrections to the ionization rates due to the inclusion of excited states as a function of the laser pulse parameters?

Methodology:

1. Define a set of laser pulse parameters (wavelength, intensity, duration) covering different ionization regimes: multiphoton limit, tunneling limit, intermediate case, simulations where the laser pulse resonantly excites atoms via multiphoton absorption and those where only virtual excitations play a role. In all cases, the ionization probabilities should not significantly exceed 0.01.
2. For each set of laser parameters in the set:
   - Run a single tRecX simulation to obtain the time-dependent probability amplitudes of all atomic states.
   - Use the script developed in Work Package 1 to extract these amplitudes and identify the relevant excited states based on the chosen criterion.
   - Utilize the modified Python code from Work Package 2 to calculate the ionization rate, incorporating the probability amplitudes of the ground state and the identified relevant excited states.
     - Gradually increase the number of excited states, starting with a calculation that includes only the ground state with the probability amplitude taken from an ab initio simulation.
   - Run the code for the same laser parameters neglecting the effect of the laser field on the neutral atom to obtain a baseline for comparison.
3. Analyze the collected ionization rate data. Visualize the results using appropriate plotting techniques.

Potential Risks and Challenges:

- **Risk:** Difficulty in identifying clear physical interpretations of the observed changes in ionization rates due to excited states.
- **Mitigation:** Compare the extreme cases where resonance-enhanced multiphoton ionization (REMPI) plays a dominant role with those where REMPI is negligible.

Expected Outcomes:

- A comprehensive dataset of ionization rates for hydrogen calculated within the SFA, both with and without considering excited atomic states, for the specified range of laser pulse parameters.
- A quantitative analysis of the corrections to the ionization rates due to the inclusion of excited states as a function of laser wavelength, intensity, and duration.
- Identification of the laser pulse regimes where the inclusion of excited states significantly affects the multiphoton ionization rate.
- Insights into the role of specific excited states in the ionization process.

2.4 Work Package 4: Documentation and Reporting

Objective: To thoroughly document the research methodology, results, and conclusions in a clear and concise manner.

Research Questions:

- How to effectively present the methodology, results, and conclusions of the research project?
- What are the key findings regarding the role of excited states in multiphoton ionization within the SFA?
- What are the limitations of the current study and potential directions for future research?

Methodology:

1. Maintain a detailed lab notebook or electronic record of all simulation parameters, code modifications, and results obtained throughout the project.
2. When preparing plots, keep well-documented Python scripts that generate the plots. This makes it easier to change plots if the input data changes, as well as to make production-quality figures for publications.
3. Write a comprehensive research report or manuscript that includes:
    - An introduction to the topic of multiphoton ionization and the role of excited states.
    - A detailed description of the theoretical framework (SFA) and the numerical methods used (tRecX).
    - A thorough explanation of the modifications made to the existing Python code.
    - A presentation and discussion of the calculated ionization rates and the effect of excited states for various laser pulse parameters.
    - A conclusion summarizing the main findings and their implications.
    - An outlook on potential future research directions.

Potential Risks and Challenges:

- **Risk:** Difficulty in organizing and presenting a large amount of data and results.
- **Mitigation:** Plan the structure of the report early in the project. Use clear and concise language and focus on the most significant findings.
- **Risk:** Potential for overlooking important details or making errors in the documentation.
- **Mitigation:** Regularly review and update the documentation throughout the project. Seek feedback from colleagues on the draft report.

Expected Outcomes:

- A well-documented research project, including simulation parameters, code modifications, and results.
- A comprehensive research report or manuscript suitable for publication in a peer-reviewed scientific journal.

3 Step 3: Overall Project Timeline

Timeline:

- **Work Package 1: Extraction of Time-Dependent Probability Amplitudes from tRecX:** 1 week
- **Work Package 2: Modification of the SFA Python Code:** 2 weeks
- **Work Package 3: Calculation of Ionization Rates and Analysis:** 7 weeks
- **Work Package 4: Documentation and Reporting:** After the first 10 weeks