What is STIRAP ?

If you are a passerby visiting this website, you may wonder what STIRAP is. „STIRAP“ stands for „stimulated Raman adiabatic passage”. It is a laser-based method for the manipulation of the distribution of population over the quantum states of an object – see below. It is used mainly in physics and chemistry but also in quantum information processing and other fields of science; technological applications are also being considered. The entire symposium “Stimulated Raman adiabatic passage in physics, chemistry and technology” (Kaiserslautern, Sept. 22 – 25, 2015) is dedicated to the discussion of related new developments and applications.

Before approaching questions regarding STIRAP we need to discuss some very basic properties of matter, in particular the quantum structure of it.

1. About quantum structure

Submicroscopic objects, such as atoms or molecules, have a quantum structure, meaning that these objects have well-defined discrete energy states. As a consequence they can absorb and release (or emit) energy only in certain well-defined discrete amounts of energy. The process of absorption and emission causes a transition between energy states of the object. In general: whenever objects are not freely moving but confined to a certain volume by forces exerted on them, they show quantum structure.

When an object is left alone (for instance when it is not irradiated by light), it will be usually found in its lowest energy state. In the language of quantum physics one says “the lowest energy state, the ground state, is populated”. When the system is in its ground state, it cannot emit energy. However it can absorb energy, e.g. in the form of a quantum of light, called a photon. When absorbing a photon the system makes a transition to a state with more energy, called an “excited state”. A system in an excited state usually releases its energy by emitting a photon. In doing so it makes a transition to a lower-lying energy state, possibly but not necessarily to the ground state. How quickly that emission process occurs after the object has been put into an excited state depends on the nature of the object and the given quantum state. One can actually not precisely define the period between excitation and emission but give only an average value, which is called the lifetime τ of the system in the given quantum state. For quantum states in atoms and molecules this lifetime τ is typically of the order of 10 ns (1 ns is a billionth of a second), but it can also be of the order of 1 μs (a millionth of a second) or even longer.

The quantum structure is also realized for larger systems, like molecules with many atoms. Then, the separation of the energy states can be very small and it may be difficult to associate a given absorption or emission process with the transition between two specific quantum states; in the language of quantum physics: it is then difficult to “resolve” the energy states.

Objects other than atoms or molecules like the light field in optical cavities or microscopic mechanical devices when cooled to very low temperatures may also reveal quantum structure.

2. Why quantum structure matters

Changing the quantum state of a system may dramatically alter its physical or chemical properties. A certain chemical reaction may not be possible when the system is in its quantum-mechanical ground state but may occur when it is in a suitable excited state. Also physical properties such as the ability to absorb light of a certain color depend on which quantum state of the system is populated. Therefore, finding schemes for the external control of the distribution of population over the quantum states has been the dream of experimentalists for many decades, in particular since the invention of the laser. Indeed, most control schemes for quantum systems rely on the interaction of the quantum system with light, in particular with laser light. This is because laser light has a specific property called "coherence", which we do not discuss here, except for stating that the frequency (color) of the light is precisely known when the laser light is coherent. The coherence of the light is essential in most schemes for controlling quantum systems. STIRAP is one of the successful and now widely applied schemes for quantum-state control.

3. What STIRAP does

We consider three states of a quantum system, see Fig. 1. In most cases, systems have many more quantum states. It is, in particular, important to note that state 2 can emit radiation (in the language of physics: it can "decay") to states other than state 1 and 3 (summarized in Fig.1 as state 4). This process is called "spontaneous emission". We assume that the quantum system is initially in state 1 (symbolized as the solid vertical bar associated with state 1). **STIRAP** aims at the ***complete*** transfer of that population into state 3 (symbolized as the dashed vertical bar associated with state 3) by coupling the quantum states 1 and 3 through radiation fields (i.e. by light) labelled P and S via a decaying state 2. The coupling via an intermediate state 2 is needed because often so-called "selection rules" do not allow the direct transition to state 3.

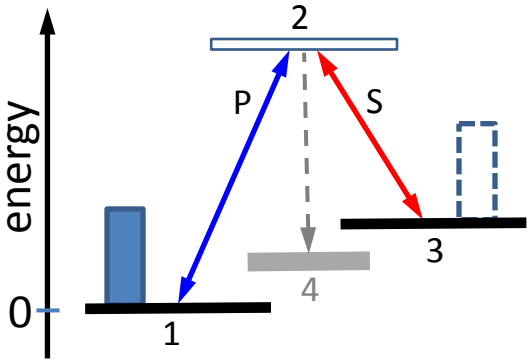


Figure 1: A set of three quantum states in the so called Lambda scheme, coupled by the radiation from two lasers, P and S. Other levels, to which state 2 can also decay, are symbolically combined as "level 4".

Figure 2 shows the evolution in time of the laser intensity and the flow of population for the STIRAP process. From the perspective of population transfer, it is very surprising that the S-field, which couples the initially unpopulated states 2 and 3, interacts with the quantum

system ***before*** the P-field does. The intensity of the P-field rises only after the intensity of S-field has reached its maximum. This is often referred to as a "counter-intuitive" sequence of pulses.

The timing of the radiative interaction is either done by controlling the time delay between laser pulses or by sending a stream of particles through spatially suitably displaced laser beams, see Section 4. Even when the energies of the photons of the fields P or S match exactly the energy differences between the respective states (in the language of quantum physics: the light fields are "on resonance" with the respective transition), level 2 is not populated. Lossless transfer of population from state 1 to state 3 occurs. In the language of quantum physics: it is the destructive interference of the two transition amplitudes which makes this process happen.

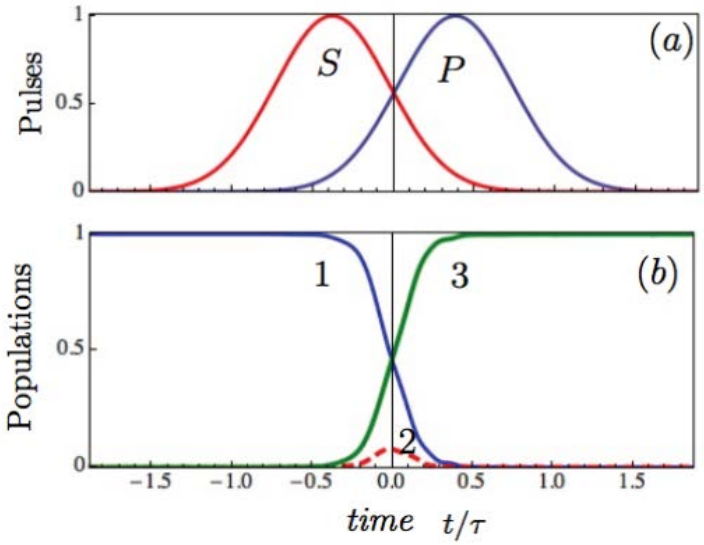


Figure 2: (a) The typical timing of the interaction of the S- and P-light with the quantum object. (b) The related evolution of the population in states 1 and 3. The population of state 2 is shown as dashed line. If STIRAP is properly implemented, the population in state 2 remains negligible and thus no loss occurs by spontaneous emission to levels.

The energy of state 3 doesn´t need to be lower than the energy of state 2, it can also be higher. In that case one speaks of a "ladder-like scheme". In that case the transfer process needs to be completed before the population of state 3 is appreciably reduced by decay.

One reason for the success of STIRAP is the robustness of the process: when STIRAP is properly implemented, the transfer efficiency remains close to unity even when the laser intensities or the time delay vary slightly. Only one condition is critical: the energy difference of the P- and S-photons must be equal to the energy difference of states 3 and 1 (in the language of quantum physics: "two-photon resonance" must be maintained).

The STIRAP scheme can be applied to any object that has a suitable quantum structure. STIRAP has also inspired similar schemes for the coupling of wave guides (i.e. the transfer of light between optical fibers or other light-guiding structures) or for transfer processes in classical systems.

4. Implementation of STIRAP

The specific timing of the interaction of the laser radiation with the quantum system can be implemented in several ways. When e.g. a sample of atoms or molecules in a gas cell is considered, STIRAP is implemented with pulsed lasers with the S-laser pulse arriving at the cell slightly ahead of the P-pulse but overlapping with it, see Fig. 2. Provided the so-called "adiabaticity criterion" is met, which essentially means that the maximum laser intensity needs to be sufficiently high and the intensities must vary smoothly with time or space, the population of state 1 is transferred entirely to state 3 while the population of state 2 remains negligibly small. If state 2 were to be populated, loss of population to other levels by spontaneous emission would occur with the consequence that population, which should be transferred to state 3, cannot reach that state.

The beam of particles (atoms or molecules) emerges from the beam source and is collimated by a pinhole, which assures that only a highly directed beam of particles reaches the S- and P-lasers. The axes of these two lasers are parallel but the S-beam is slightly displaced to a position upstream of the P-beam, still overlapping the latter. As the particles fly through the laser beams, they are first in contact with the S-laser, next with both lasers and finally with the P-laser only. The timing of the interaction is as shown in Fig. 2 a.

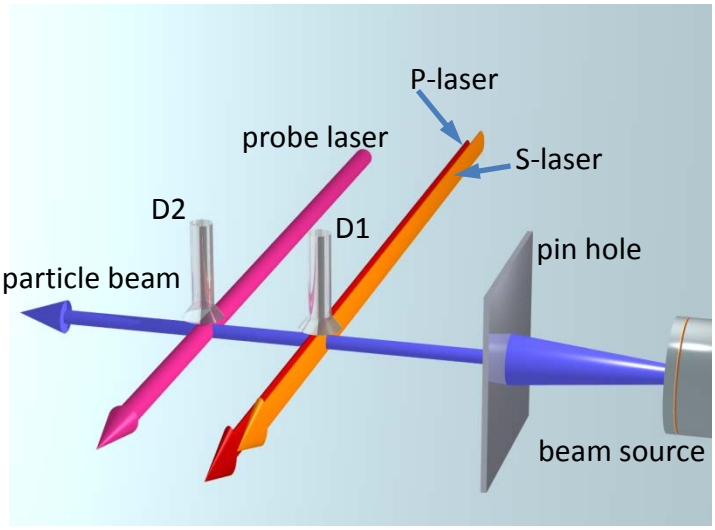


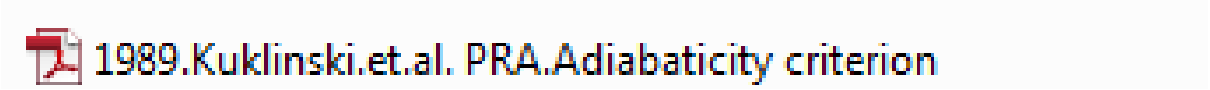
Figure 3: A typical setup for the implementation of STIRAP with continuous lasers and an atomic or molecular beam. The transfer occurs in the region were the S- and P-laser beams overlap. The detector D1 monitors fluorescence, if any, emitted from state 2 in that region. Detector 2 monitors the population of state 3 by laser-induced fluorescence.

Detector D1 monitors any light emitted from state 2 in the transfer region. When the conditions for efficient transfer by STIRAP are met, the signal at D1 is negligibly small. The success of the transfer is monitored by detector D2 further downstream. It detects the fluorescence induced by the probe laser. That fluorescence is a measure of the population in state 3.

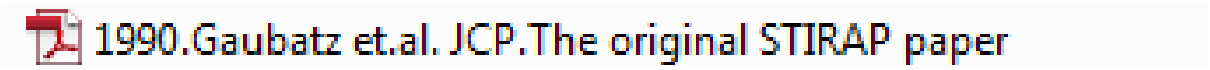
5. Further Reading

The interested reader will find more details in the publications listed below (next page).

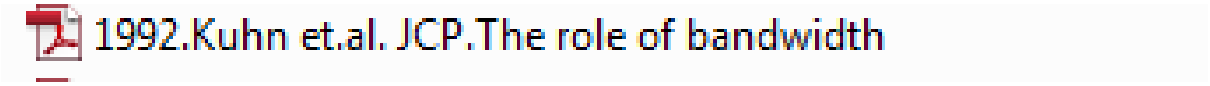
The paper which discusses first some conditions that need to be met for successful implementation of STIRAP. (Kuklinski, Gaubatz, Hioe, and Bergmann: “Adiabatic population transfer in a three-level system driven by delayed laser pulses”, Phys. Rev. A 40, 6741, 1989):



The “original” STIRAP paper, with the basic theory and a convincing set of experimental data. (Gaubatz, Rudecki, Schiemann, and Bergmann: “Population transfer between molecular levels by stimulated Raman scattering with partially overlapping pulses: A new concept and experimental results”, J. Chem. Phys. 92, 5363, 1990):



Discussion of the consequence of phase fluctuations for STIRAP. (Kuhn, Schiemann, He, Coulston, Warren, and Bergmann: “Population transfer by stimulated Raman scattering with delayed pulses using spectrally broad light”, J. Chem. Phys. 96, 4215, 1992):

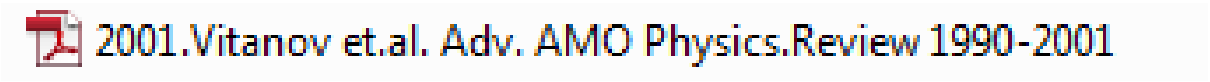


STIRAP shown for the SO_2 molecules, to date still the largest molecule for which STIRAP has been shown experimentally to be doable. (Halfmann and Bergmann: “Coherent population transfer and dark resonances in SO_2”, J. Chem. Phys. 104, 7068, 1996):

STIRAP shown for a multi-level system with level spacing externally controlled by a magnetic field. (Martin, Shore and Bergmann: “Coherent population transfer in multilevel systems with magnetic sublevels III. Experimental results”, Phys. Rev. A 54, 1556, 1996):

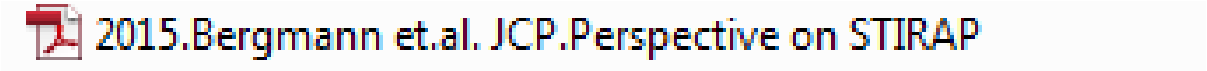
A highly cited review, serving as an introduction to STIRAP. (Bergmann, Theuer, and Shore: “Coherent population transfer among quantum states of atoms and molecules”, Rev. Mod. Phys. 70, 1003, 1998)

A comprehensive review of the field until the year 2001. (Vitanov, Fleischhauer, Shore, and Bergmann: "Coherent manipulation of atoms and molecules by sequential pulses", Adv. in Atomic, Molecular and Optical Physics, 46, 55, 2001):



A review of work showing how to prepare controlled superposition states with STIRAP. (Vewinger, Shore, and Bergmann: "Superposition of degenerate quantum states: preparation and detection in atomics beams", Adv. in Atomic, Molecular and Optical Physics, 58, 113, 2010):

A most recent discussion of the motivation, development, status and possible future developments of STIRAP focusing on examples, not a comprehensive review. (Bergmann, Vitanov, and Shore: "Perspective: Stimulated Raman adiabatic passage: The status after 25 years", J. Chem. Phys. 142, 170901, 2015):



Discussion of the „pre-history" of STIRAP, showing what elements were discussed before 1990 and how - including the concept of dark states in the papers by W. Lamb from the early 1950s (Shore: "Pre-history of the concepts underlying stimulated Raman adiabatic passage (STIRAP)", Acta Physica Slovaca 63, 2013 [appeared in December 2014]):